

Neutron Backscattering

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Abstract

We review the status of neutron backscattering spectrometers on reactors and spallation sources including the most recent instrument developments. After setting the stage by looking briefly at the history of backscattering instruments, we review the current state-of-the-art in backscattering spectrometers leading to the most recent projects. In particular, we discuss the developments at reactors which have led to the classic type of backscattering spectrometers. These instruments typically use cold neutrons along with crystalline monochromators and large crystalline analyzers both of which diffract neutrons very nearly in exact backscattering thereby providing energy resolutions < 1 μeV . More recently, powerful spallation pulsed neutron sources have been built allowing inverted-geometry instruments in which a pulsed, broad-energy band of neutrons strikes the sample. The final neutron energy is then specified by a large crystalline analyzer arranged so that the neutrons are diffracted near backscattering providing a somewhat relaxed energy resolution from that of the typical reactor-based instruments. Both types of backscattering spectrometers are essential members of the basic instrument suite for high energy resolution spectroscopy.

Introduction

Neutron **BackScattering Spectroscopy** (BSS) was invented in the 1960's to improve the energy resolution of neutron instrumentation into the sub- μeV region – a range unattainable with conventional triple-axis or time-of-flight spectrometers [Maier-Leibnitz 1966][Alefeld 1967][Alefeld 1969]. In fact, the energy resolution of a triple-axis instrument is limited by the beam divergence, crystal quality and, most-importantly, the Bragg angle. In fact, a backscattering spectrometer can be considered an extreme case of a triple-axis spectrometer where the resolution limitation imposed by these crystal optics is overcome. In particular, as its name suggests, “backscattering” refers to neutrons scattered from the monochromator and analyzer crystals through 180° . This allows these instruments to typically provide an energy resolution with a full-width at half-maximum FWHM on the order of $1 \mu\text{eV}$, corresponding to a time scale of ns. This range encompasses important dynamics in materials ranging from polymers and biomaterials to porous solids and energy storage materials to magnetic systems. It is important to note that the term backscattering refers only to the neutron optical components which determine the resolution and not to scattering from the sample. Thus the sample scattering can be investigated through a normal range of scattering angles giving scientists access to a significant range of momentum transfers and therefore length scales of the observed motion.

The energy width provided by Bragg diffraction from a crystal can be obtained simply by differentiating Bragg's law to yield:

$$\Delta E = 2E (\cot\Theta \Delta\Theta + \Delta d/d) \quad \text{eq. (1)}$$

where E is the energy of the neutrons, Θ is the Bragg angle, and d is the lattice spacing of the crystal. The $\Delta d/d$ term is usually ignored but as we will see is important for backscattering. This equation suggests that the resolution provided by crystal optics can be improved by decreasing $\Delta\Theta$ by using perfect crystal monochromators instead of mosaic crystals or by limiting the divergence provided to the crystals using collimators. Either of these measures leads to extremely low count rates that are unsuitable for most experiments. Alternatively one can minimize $\cot\Theta$ by using a Bragg angle of nearly 90° meaning that the neutron will be backscattered from the monochromator. Because the coupling between divergence and final energy becomes negligible in backscattering this approach also allows one to increase the divergence and thus promises larger count rates. However, it is more difficult to achieve in practice. It was the group of Maier-Leibnitz who built the first realistic neutron spectrometer operating at Bragg angles very close to 90° in Garching near Munich [Alefeld 1967][Alefeld 1969][Birri 1971].

To further address the inherently low intensity for high resolution spectrometers, this group designed and built a large spherical backscattering analyzer thereby increasing the solid angle of analysis [BS-website]. This relaxes the Q -resolution which is acceptable for scattering laws with weak momentum transfer dependence. Fortunately,

this is usually the case for inelastic scattering particularly for polycrystalline or amorphous samples or for incoherent scattering.

The Garching group also devised ways to vary the energy of the neutrons which are incident on the sample without scanning the Bragg angle which would degrade the energy resolution by moving the monochromator or analyzer away from backscattering. They showed that maintaining the backscattering condition is possible by either rapidly moving the monochromator parallel to the incident beam (a Doppler monochromator) or by changing the lattice spacing of the monochromator using thermal expansion via a finely controlled temperature stage. Their first BSS was soon followed by spectrometers in Jülich, Germany [Alefeld 1972] and at the Institut Laue-Langevin, Grenoble, France [Heidemann 1978][IN10 website] (IN10). These cold neutron spectrometers used Doppler drives to change the incident energy. In contrast, a variant of IN10 - IN10B [Cook 1992] and the only thermal neutron BSS IN13 [Heidemann 1977][IN13 website] use thermal expansion of a heated monochromator. Both IN10/B and IN13 are still in operation. As pulsed neutron sources were commissioned, the primary spectrometers of BSSs were adapted to use the time structure of the source. These TOF-backscattering spectrometers which are inverted geometry time-of-flight spectrometer [Carlile 1992] where the energy is analyzed by a large set of analyzer crystals set close to backscattering are discussed later in this chapter

The energy resolution near backscattering from perfect crystals

Eq. 1 showed that the factors contributing to the energy resolution of BSSs can be divided into an angular term and another proportional to Δd . The angular term can be expanded around a Bragg angle of 90° yielding:

$$\Delta E/(2E) \approx (\delta\theta + \Delta\theta/2)^2/2 + \Delta d/d \quad \text{eq.(2)}$$

The first term in eq.(2) is an approximation valid near Bragg angles of 90° , where one can express the deviation from backscattering as contributions from the beam divergence, given by $\Delta\theta$, and deviation from a 90° Bragg angle, given by $\delta\theta$ (see Fig.1a). Note that both $\delta\theta$ and $\Delta\theta$ contribute only in second order. Also, in backscattering, the angular deviations $\Delta\theta$ and $\delta\theta$ only provide larger energies (corresponding to shorter wavelengths) compared to the nominal one. By comparing the shortest and longest wave-vectors \mathbf{k}_{\min} , \mathbf{k}_{\max} ($\mathbf{k} = 2\pi/\lambda$) scattered under the conditions illustrated in Fig.1b and noting that $\Delta\theta$ refers to the full divergence, one finds the factor 1/2 in the this term in eq.(2) [Birr 1971].

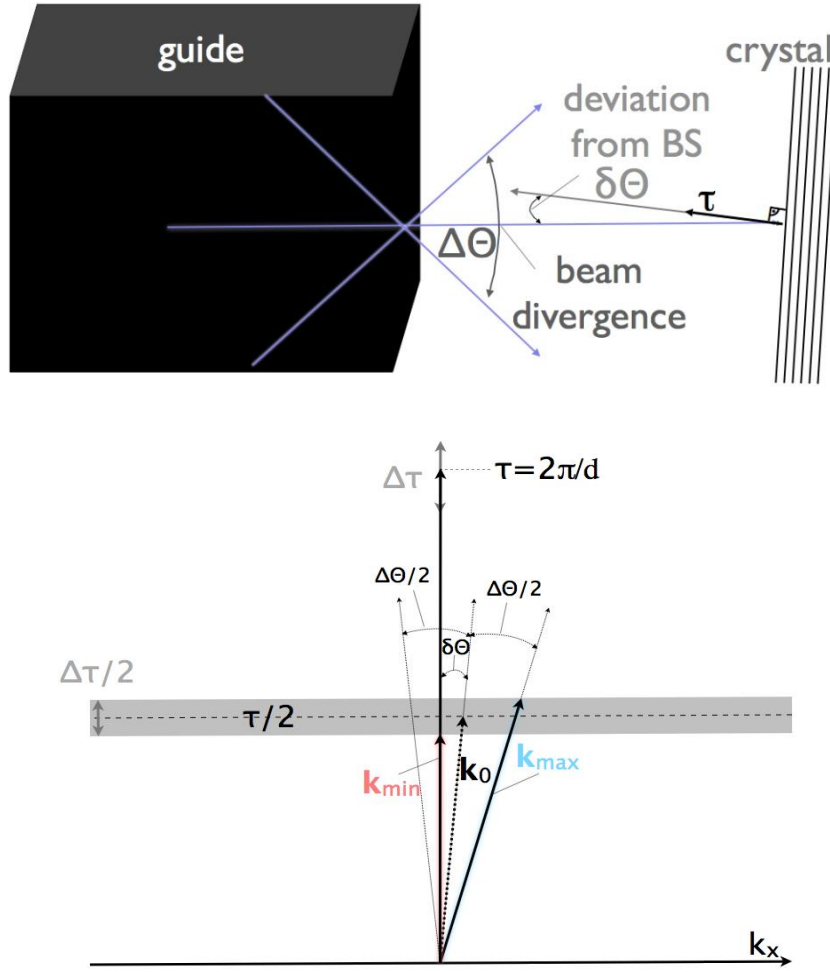


Fig.1: a) Illustration of beam divergence $\Delta\Theta$ and deviation from backscattering $\delta\theta$ for $\delta\theta < \Delta\Theta$; the beam divergence is centered around the deviation angle $\delta\theta$. For a neutron guide with ^{nat}Ni and for the Si(111) backscattering wavelength 6.27\AA the maximum deviation from backscattering due to divergence is equal the critical angle, thus $\Delta\Theta/2 \sim 0.627^\circ$, therefore contributing near backscattering about $0.25\mu\text{eV}$ to the energy resolution (see eq.2). b) reciprocal space consideration for estimating the angular deviation contribution to the resolution near backscattering for $\delta\theta < \Delta\Theta$. The reciprocal lattice vector τ has an uncertainty due to primary extinction and lattice strains which determines which wave-vectors \mathbf{k} are accepted out of the divergent beam.

The last term in eq.(2) is the Darwin width, which refers to the uncertainty in the d-spacing for perfect crystals due to primary extinction. Thus even in the case of “perfect” backscattering the resolution is not perfect. Rather the Darwin width of the Bragg reflections from the monochromator and analyzer crystals sets the best achievable energy resolution which e.g. for perfect Si(111) crystals contributes about $0.077\mu\text{eV}$ to ΔE (see Table1). Note that the Darwin width increases linearly with the structure factor of the Bragg reflection [BS-website] . Thus the ultimate resolution limit is improved for the GaAs(002) reflection compared to Si(111) because the structure

factor for GaAs(002) is proportional to the small difference in the scattering lengths of Ga and As.

crystal plane	$\Delta\tau/\tau$ (10 ⁻⁵)	ΔE_{ext} (μeV)	$\lambda(\text{\AA})$ for $\Theta=90^\circ$
Si(111)	1.86	0.08	6.27
Si(311)	0.51	0.08	3.27
CaF ₂ (111)	1.52	0.06	6.31
CaF ₂ (422)	0.54	0.18	2.23
GaAs(400)	0.75	0.15	2.83
GaAs(200)	0.16	0.01	5.65
Graphite(002)	12	0.44	6.7

Table 1: Best possible energy resolution for some perfect crystals suitable for backscattering, which in dynamical scattering theory is given by the uncertainty in the reciprocal lattice vector $\tau = 2\pi/d$ corresponding to the reflection used, equivalent to the uncertainty in lattice spacing d (first term of eq.1). It is crucial to note that the resolution is determined by the structure factor [BSS website].

Trading energy resolution for intensity - Less perfect crystals

The count rate of a backscattering spectrometer can be enhanced with little loss of resolution by matching the contribution from the $\Delta d/d$ term in eq. 2 to that of the angular terms through crystal engineering. This is most often done by intentionally deforming the crystals by bowing them when they are affixed to the carefully machined backing plates. For spherically bowed crystals,

$$\Delta d/d = (\Delta d/d)_{\text{Darwin}} + P_{\text{eff}}(t/R_c) \quad \text{eq.(3)}$$

where $(\Delta d/d)_{\text{Darwin}} \approx 1.86 \times 10^{-5}$ is the Darwin peak width, $P_{\text{eff}} = 0.44$ is Poisson's ratio for this crystal orientation, t is the thickness of the crystal (between 250-2000 μm) and R_c is the radius of curvature. Thus by bending the crystals, an increased number of lattice planes contributes to the reflection, sacrificing resolution for intensity. This technique was applied to the early IN10 analyzers, which had a “polished” (unstrained) and ‘unpolished’ analyzer setup. In fact “unpolished” referred to small hexagonal crystals (~10mm side length) with a lapped waver surface which were glued under deformation onto the spherical support of the analyzers. It is important to note that measurements indicate that the additional strain imposed by bowing is partially relieved due to the finite lateral size of the crystals. Thus eq. 3 underestimates the required thickness to achieve a desired $\Delta d/d$. This and the more inhomogeneous strain distribution of small crystals led to the use of large deformed wafers (100-120 mm diameter) on all later BSS.

The energy resolution of a complete backscattering spectrometer

The above resolution considerations are sufficient to understand the design of a BSS. In fact, all geometrical contributions to the energy resolution like beam divergence, beam size, sample size, crystal size and detector size are taken into account by the angular contributions in eq.(2) and the crystal term by the $(\Delta d/d)$ term. The total energy resolution of any BSS is then the convolution of the contributions in eq.(2), arising from the monochromator in the primary spectrometer and from the analyzers in the secondary spectrometer:

$$\delta E = (\delta E_p^2 + \delta E_s^2)^{1/2} \quad \text{eq.(4)}$$

As we will see later, the resolution of the primary spectrometer is given by a flight time resolution contribution for a TOF-Backscattering instrument.

The first generation of reactor backscattering spectrometers

Fig. 2 shows a sketch of IN10 which is representative of the first backscattering spectrometers [IN10-website]. The arrangement of a perfect backscattering geometry is geometrically difficult, a fact which is reflected in the design of both the primary and secondary spectrometers. The IN10 monochromator is placed at the end of a natural Ni guide, which deflects the beam towards a graphite deflector crystal placed above the neutron beam about 6 m upstream from the monochromator. This geometry means that the Bragg angle deviates slightly from backscattering with the angular deviation chosen to be smaller than the critical angle of the neutron guide. The resolution contribution (ΔE) from the monochromator system due to the deviation from backscattering ($\delta\theta$) is estimated to be $\approx 0.1 \mu\text{eV}$ and from the guide divergence ($\Delta\theta$) $\approx 0.25 \mu\text{eV}$. These should be compared to the extinction contribution (from the $\Delta d/d$ term) of $0.08 \mu\text{eV}$ for Si(111).

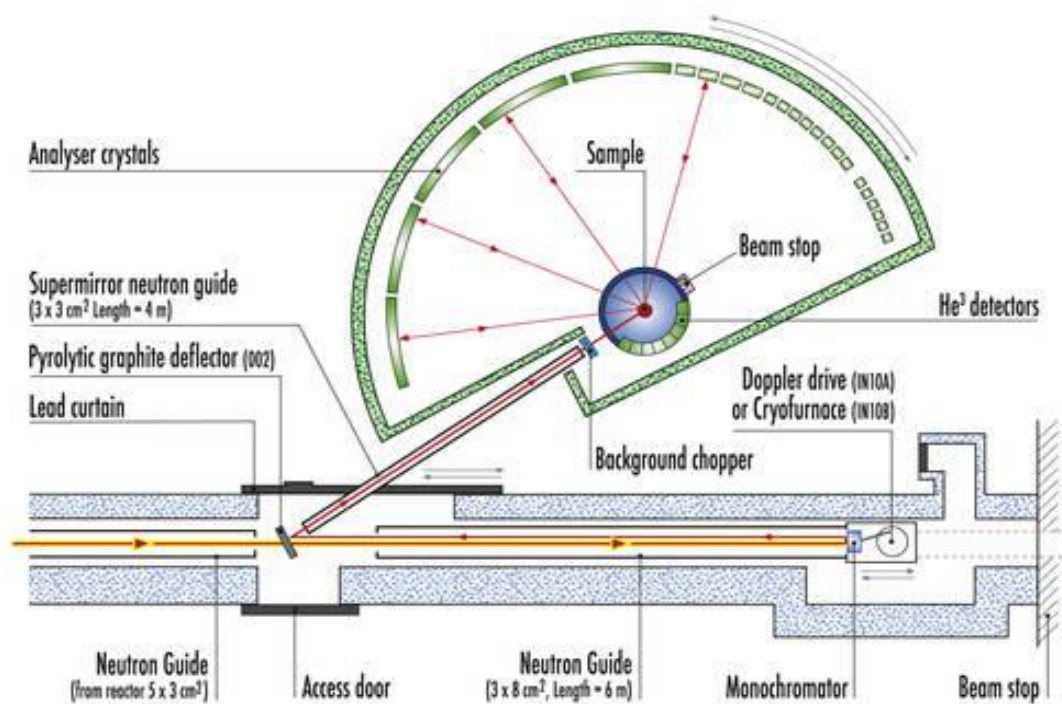


Fig.2: IN10, representative of a first generation backscattering instrument [IN10-website].

The monochromatic beam is sent from the graphite deflector to the sample, passing a chopper with 50% duty cycle. As the detectors must have a direct view of the sample for the analyzer to be in “exact” backscattering, this chopper is necessary to exclude those neutrons which scatter directly from the sample into the detectors without striking the analyzer. For this purpose, the detector is electronically gated with the chopper phase so that the detectors are inactive when neutrons strike the sample. The neutrons which are scattered from the sample travel to a large, spherically shaped analyzer composed of perfect single crystals, normally of the same kind and orientation as the monochromator, selects a fixed final energy (2080 μeV for Si (111)). Most importantly, to provide resolution which matches that of the primary spectrometer, the analyzers are aligned in perfect backscattering ($\delta\theta = 0$). This introduces another geometrical difficulty which is solved by requiring that the detected neutrons pass through the sample a second time. Of necessity, this implies the possibility of additional scattering. Fortunately this induces only a very small correction. This fortuitous situation occurs because most neutron scattering is elastic or nearly so and the distance between the sample and the detectors is small. Thus there is only a negligible shift in the apparent energy of any twice scattered neutrons which reach the detectors. This means that the correction primarily appears in the less important Q -dependence of the scattering. Furthermore, due to the typical 10% probability of scattering by the sample and the fact that scattering goes into 4π sr, the number of these double scattered neutrons seen by the rather limited solid angle of a detector is small.

As we have just seen, the neutrons which scatter from the sample to the analyzer to the detector have a precisely known fixed final energy imposed by the analyzer. All that remains to determine the energy transferred to the sample is to ascertain the initial energy of the detected neutrons. If the incident energy is changed by varying the temperature of the monochromator, then with a sufficiently slow temperature variation, the neutron energy can be determined simply by the temperature of the monochromator (and thus the d-spacing) at the time the neutrons are detected. For Doppler-equipped spectrometers, the incident neutron energy varies with the instantaneous speed of the monochromator. Thus the detected neutrons must be related to the Doppler speed at an earlier time. This is possible because both the neutron energy and hence its velocity and the flight distance between the monochromator and the detector are well-known. Therefore it's a simple matter to relate the time at which the neutron is detected to the time it left the monochromator which is directly related to the initial energy of the neutron. The Doppler frequency is usually lower than the chopper frequency and not phase related.

Trading Q-resolution for intensity - Focusing

The first reactor BSS example IN10 was placed on a natural Ni-guide and employed a flat monochromator meaning that the guide divergence presented a non-negligible contribution to the energy resolution of the monochromator system. Moreover the Q-resolution provided by the primary spectrometer was much better than that of the secondary spectrometer. Thus the developments in neutron optics, which deliver high neutron flux based on focusing and greatly improved guide coatings provided an opportunity to increase the angular divergence, and therefore the flux of neutrons, delivered to the sample. Moreover, the increase in count rate would be at the expense of the rather unimportant Q-resolution provided by the primary spectrometer. The use of enhanced focusing optics requires replacing the flat, perfect crystal monochromator with a primary spectrometer having a spherical geometry similar to that developed for the secondary spectrometer of IN10 [Alefeld 1992][Magerl 1992]. The first backscattering instrument to profit from these innovations in neutron optics was IN16 a second generation instrument at the ILL [Frick 1997][Frick 2001]. (Fig. 3)

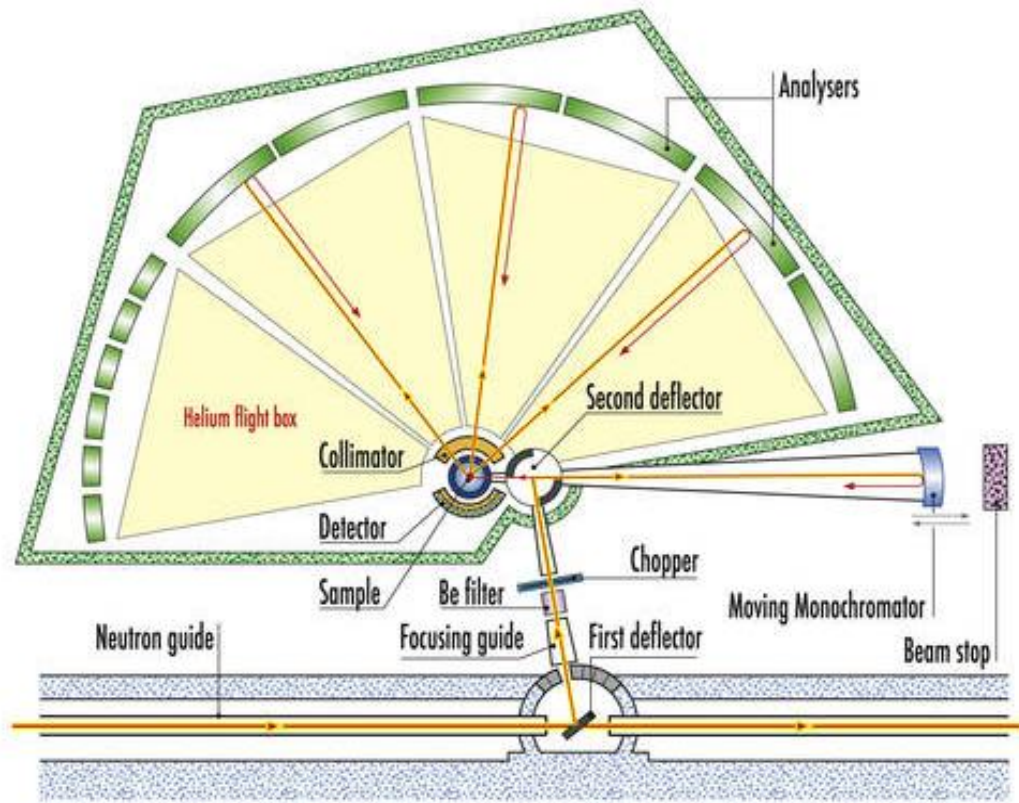


Fig.3: Schematic view of IN16. A double monochromator-guide focusing optics pre-selects a wavelength band for the backscattering monochromator. The second deflector takes also the role of a chopper [IN16-website].

IN16 uses a combination of a vertically focusing, pyrolytic-graphite neutron deflector placed in the primary guide along with a tapered neutron guide to extract a suitable neutron beam and focus it to a small high intensity beam albeit one with increased angular divergence. This deflector consists of three PG(002) horizontally inclined crystals having a mosaic of 0.5° each, thereby introducing an artificial horizontal mosaic which matches the divergence provided by a ^{58}Ni guide while not significantly increasing the vertical divergence. The beam which exits the tapered guide impinges on a second deflector, with the two deflectors arranged in the standard double monochromator focusing geometry. Starting from the second flat PG(002) deflector, the area of the now divergent beam greatly expands before it hits a large spherical monochromator in backscattering geometry. If the focal point created by the optics of deflector-guide combination can be imaged onto the sample by a backscattering monochromator, this design allows the divergence of the incident beam to be quite large without degrading the energy resolution. This means that the beam size at the focal point must be small with respect to the distance between the focal point and the backscattering monochromator. The divergence contribution to the energy resolution in such a geometry can be estimated from eq.(2) - for a spot size of $22 \times 22 \text{ mm}^2$ and a distance of 2 m to the spherical monochromator, the divergence contribution to the energy resolution is $\Delta E \approx 0.13 \text{ } \mu\text{eV}$, a value reasonably close to the extinction

contribution. Unfortunately, deviations from backscattering induced by an ill defined focal point, monochromator displacement, fixed monochromator radius and finite crystal size all degrade this “ideal” value.

In order to remain close to backscattering the second deflector on IN16 is mounted on the 50% duty cycle chopper. The chopper speed is matched to the flight time of the neutrons from the chopper to the monochromator and back. A background chopper prevents neutrons from entering the secondary spectrometer during the opening time of the deflector chopper and a cooled Be-filter suppresses the higher order scattering from the first deflector. The secondary spectrometer layout is similar to IN10, but IN16 has a larger analyzer radius (2 m compared to 1.5 m), larger solid angle coverage, a multi-tube detector assembly, and a diffraction bank below the analyzer area which allows for monitoring the structure of the material being studied. More importantly, IN16 has both a high energy resolution Si(111) configuration with a spherical array of $4 \times 4 \text{ mm}^2$ crystals ($\Delta E \approx 0.4 \text{ } \mu\text{eV}$) and a low resolution Si(111) configuration of large deformed 0.7 mm thick wafers ($\Delta E \approx 0.85 \text{ } \mu\text{eV}$) as well as a Si(311) configuration which allows to access large Q-range ($3.7 \text{ } \text{\AA}^{-1}$ compared to $1.9 \text{ } \text{\AA}^{-1}$ for Si(111)). Unfortunately, the chopper is not optimized for the short Si(311) wavelength leading to long counting times. IN16 is a particularly flexible instrument because the secondary spectrometer is under air and the analyzers and spectrometer are on air pads, which allows relatively rapid configuration changes.

Trading Q-resolution for intensity - Phase Space Transformation

With the backscattering spectrometer HFBS at the National Institute of Standards and Technology (NIST) [HFBS-website] [Meyer 2003], the recent commissioned SPHERES of the Juelich Center for Neutron Scattering (JCNS) at the 'Forschungs Reaktor Muenchen', FRMII [SPHERES-website] and the ongoing construction of IN16B a new generation of BSS is introduced. These 3rd generation spectrometers are similar and further increase the incident beam divergence using the “phase space transformation” (PST), first proposed by Schelten and Alefeld [Schelten 1984] This device requires mounting crystals on a chopper much as is done on the second deflector of IN16, but with a crystal speed at least 3x faster. More importantly, it also requires an end guide position.

As the PST effect has been the subject of many publications [Schelten 1984][Gehring 1997][Kirstein 1999][Kirstein 2000][Meyer 2003][Hennig 2009] we will only describe it briefly here. The main purpose of a PST is to offer the optimal phase space to the moving backscattering monochromator. Consider a well-collimated neutron beam such as that provided by a neutron guide. When such a beam diffracts from a stationary mosaic crystal, the phase space element is transformed into the concave-shaped element shown in the top panel of Fig. 4. Neutron energies are not changed in this process; rather the energies are sorted in angle as indicated by the colors - higher energy neutrons diffracting at lower angles. Note that the energy of many neutrons fall outside the range accepted by the backscattering monochromator, a range indicated by the parallel black lines.

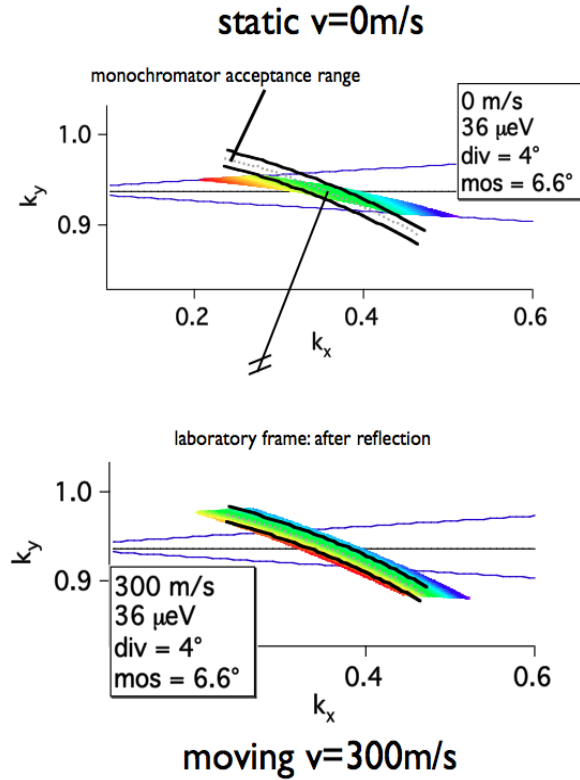


Fig. 4: Section of reciprocal space showing the result from phase space calculations of the reflection of a divergent neutron beam (4°) with wide wavelength spread. The colored points refer to neutrons which were reflected by the graphite PG(002) mosaic crystal planes. The circular black parallel curves describe the acceptance of a spherical backscattering monochromator moving at its extreme Doppler velocity corresponding to $\pm 36 \mu\text{eV}$. When the area of colored phase space in the acceptance range of the Doppler monochromator is maximized, the intensity is optimum. In the upper figure the mosaic crystal is at rest, while in the lower figure it is moving in $-\mathbf{k}_x$ direction perpendicular to the reciprocal lattice vector (parallel \mathbf{k}_y direction).

The situation is different, however, if the crystal is moving. If the motion is antiparallel to the projection of the direction of the incoming neutrons onto the Bragg planes of the mosaic crystal (*i.e.* to the left in Fig. 4), the concave element rotates in phase space. More importantly, the rotation is such that shorter wave vectors become elongated while longer wave vectors are shortened. When the speed is well-chosen, this phase space transformation results in the situation shown in the bottom panel. Here many more of the neutrons have energies lying within the band defined by the parallel solid lines. Moreover these neutrons subtend a considerably larger angle. Thus the PST converts a wide wavelength band with lower divergence into a narrower wavelength band with larger divergence – the transformation is from ‘white to wide’ [Schelten 1984] in agreement with Liouville’s theorem. The necessary conditions for employing a PST are: i) the availability of a wide wavelength band (typically this requires an end-guide

position), ii) the mosaic of the PST crystal has to be large enough to accept the wide wavelength band as well as the divergence provided to the PST and iii) the deflector (PST) crystal has to move with a speed of 250-300 m/s perpendicular to the scattering plane and the reciprocal lattice vector of the reflection used.

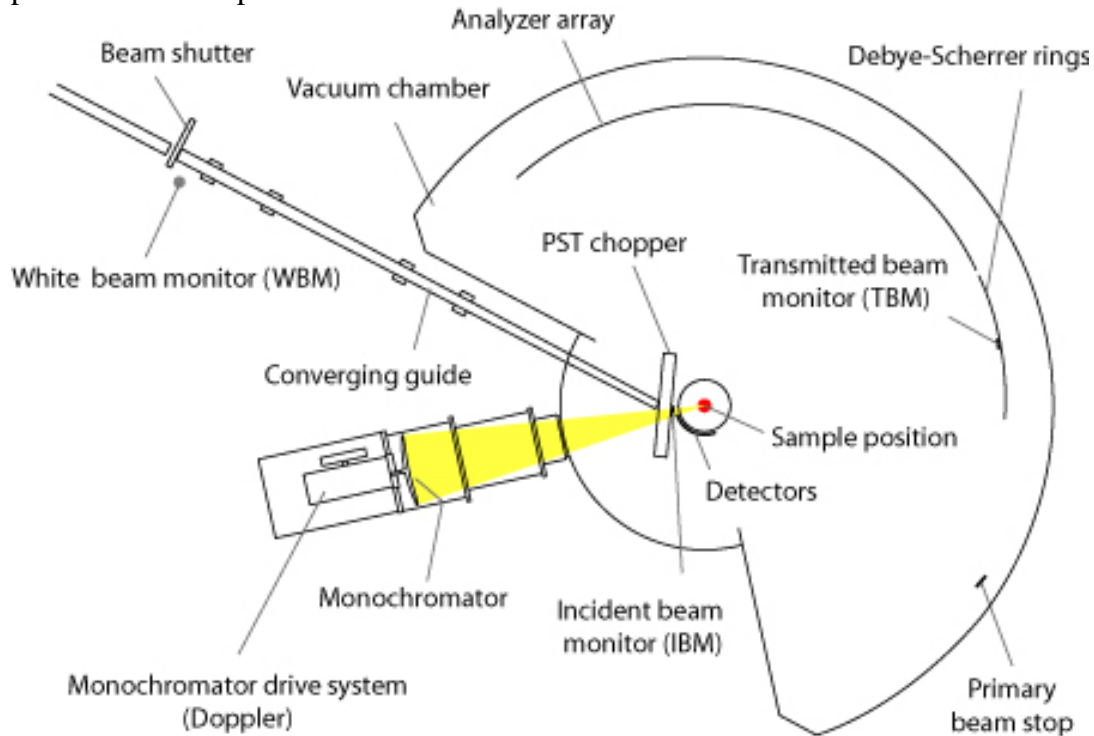


Fig.5 Schematic layout of the HFBS, NIST

The first spectrometer to utilize a PST chopper was HFBS at NIST [Meyer 2003] [Gehring 1997][HFBS-website] (Fig.5). (A very similar PST spectrometer, SPHERES, built by JCNS recently began operations at the FRM-II Munich.) The primary spectrometer utilizes the spherical focusing geometry developed for IN16. However, rather than using an in-guide, stationary deflector which would substantially limit the wavelength band available to the PST, the guide transports the neutrons directly to the PST chopper which deflects the neutrons to the monochromator. The neutrons are then backscattered from the monochromator, passing through an opening in the PST chopper to the sample. Note that the linear speed of the crystals which are mounted on the chopper is 250 m/s. Compare this to the approximately 80 m/s crystal speed on the second IN16 deflector and the technical challenge of building such a chopper becomes clear. The additional wavelength band and the much higher crystal speeds combine to provide a measured gain a factor 4 when the crystals are moving at the design speed compared to when they are stationary.

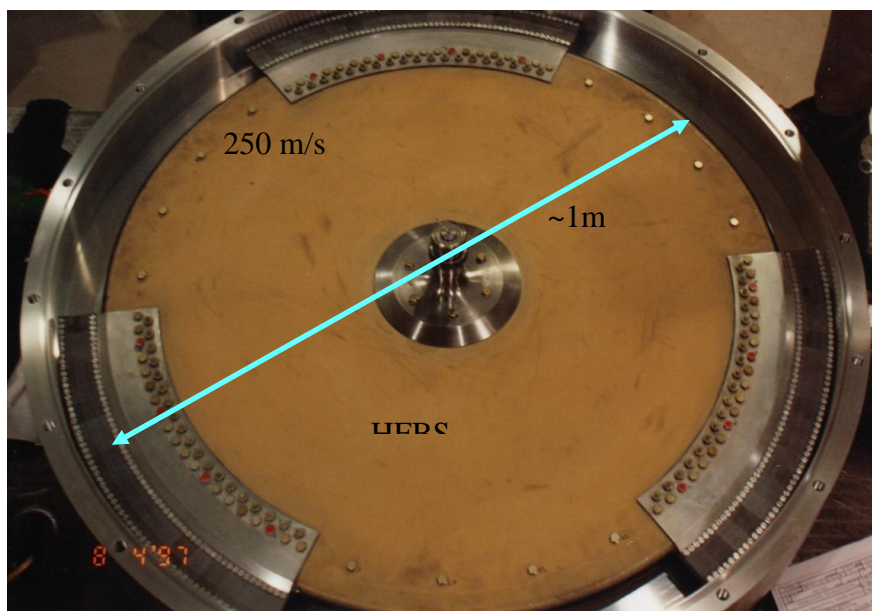


Fig. 6 shows the PST disk and the graphite cassettes at its outer border, packed into cassettes for increasing the mechanical stability.

The disk (Fig. 6) consists of three segments each enclosing 60 graphite crystals having a nominal mosaic of 2.5° . Much like the in-guide deflector on IN16, an artificial mosaic is produced by stacking three PG(002) so that they are horizontally inclined with respect to each other thus introducing an artificial horizontal mosaic of 7.5° . Thus the beam leaves the chopper with a divergence of 15° seen in the yellow band of Fig. 6. Clearly the Q-resolution provided by HFBS is quite relaxed. Due to the stacking arrangement of the graphite crystals in the chopper, the vertical mosaic is only 2.5° thereby limiting the vertical beam divergence and making it easier to focus the neutrons back to the sample position. In addition to the necessity for the crystals to achieve a linear speed of 250 m/s, the rotational frequency (and thus the chopper diameter) are set by the requirement that the PST chopper must have opened in the time it takes neutrons to fly from the chopper to the monochromator and back. The chopper on HFBS is designed so that the 1 m diameter disk turns 180° in this time.

In the similar spectrometer SPHERES, the PST disk has a diameter of about 1.3 m providing a crystal speed of 300 m/s (although it is currently operating at 1/3 of this design) which was originally believed to be optimum [Schelten 1984]. However the maximum gain depends only weakly on the speed for a reasonable mosaic of 5° to 10° and for the divergence that modern neutron optics can provide. More recent simulations suggest that for the geometry of the new IN16B being built at the ILL, the maximum gain is achieved for 250 m/s [Hennig 2009].

A disadvantage of the PST concept is that it is necessary to bring a wide wavelength band (meaning a high flux of neutrons) into the secondary spectrometer. In fact the beam stop for this is located only about 20-40cm from the detectors. Therefore it is difficult to achieve a good signal to background ratio. The HFBS instrument employs a velocity selector which limits the wavelength band and thereby reduces the background

by a factor of 8 with only a 15% reduction in the signal. The background is further reduced by reducing the scattering from air in the secondary spectrometer by either building a vacuum chamber (HFBS) or by filling the secondary spectrometer with Ar (SPHERES). Finally, like on IN16, one can incorporate a background suppression chopper which prevents neutrons from entering the chamber from the guide when neutrons are striking the sample (SPHERES). A combination of measurements and simulations suggest that this device further reduces the background by a factor of 2, albeit with a small decrease in the intensity [Garcia Sakai 2008],

Improving the dynamic range

Since the earliest days of backscattering spectrometers, efforts have been made to increase the energy range accessible by these instruments. This desire led to the development of offset monochromators in which the monochromator has a slightly different d-spacing from the analyzer. Perhaps the best example of this is the development of SiGe alloys at the ILL for use as a backscattering monochromator [Magerl 1990]. Found to be of more utility, was adjusting the d-spacing with thermal expansion through the use of cryofurnaces. These devices, which are in regular use at IN13 and IN10B, produce a well-controlled and exquisitely uniform temperature environment for the monochromator [Cook 1992]. However, the advent of large focusing monochromators made this approach technically daunting. Thus more recent efforts have concentrated on the development of Doppler monochromators capable of driving larger loads at higher speeds. Because the PST approach presents a larger energy band to the monochromator than is available when using an in-guide deflector, the first significant development along this line was made for HFBS. This spectrometer employs a counterbalanced mechanical device which is capable of providing speeds corresponding to energy transfers of $\pm 50 \mu\text{eV}$ although in practice vibrational resonances limit the useful range to $\pm 36 \mu\text{eV}$. An alternative approach taken at SPHERES is the use of linear motors, moving a carbon fiber piston and monochromator on very thin air cushions [Doppler AEROLAS]. The linear motor Doppler drive allows users a great deal of flexibility in choosing a velocity profile and amplitude. Thus, in principle, one can distribute the neutron energies over the range of interest in a way which produces the most information in the shortest amount of time. In order to suppress vibrations, it is mounted on a heavy granite block. The same drive with improved control is now installed at IN16.

Ongoing backscattering projects

At ILL a new BSS, IN16B, is under construction (Fig.7) with the aim to combine all progress achieved on IN16, HFBS and SPHERES [Frick 2006]. IN16B will be placed at the end of a very long ballistic cold neutron guide where it profits from modern focusing optics and PST-effect, providing a high count rate and a wider dynamic range [Bordallo 2009]. In addition, it will be able to sweep to an IN16-like side position, which guarantees good background conditions. Similar to HFBS, IN16B will have vacuum in the secondary spectrometer and in all flight path. Flexibility is maintained for extending to high Q with Si(311) crystals and to high energy resolution with

GaAs(002). Operation as an inverted time-of-flight-backscattering option has been designed and is currently under review.

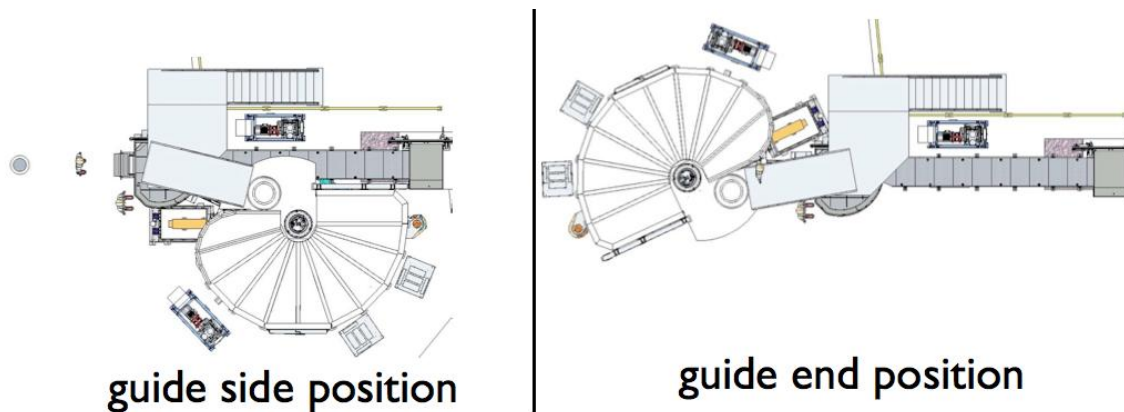


Fig.7 The design of IN16B allows it to sweep between a side position to work in a IN16-like low background mode and a high-flux guide-end position which employs a PST chopper.

The PST disc of IN16B with a diameter of 66cm is planned to be more compact than the PST of HFBS and SPHERES. Like on these spectrometers 3 graphite crystals will be assembled in a cassette, with a slight horizontal inclination angle of 2.2° between them to obtain an effective horizontal mosaic of about 6.6° . IN16B assembles single cassettes on a chopper wheel with 2 (rather than 3) windows allowing for a more compact disc. The technical challenge is packaging these rather fragile graphite crystals in a way that allows to rotate them at 243m/s with a corresponding centrifugal acceleration of $1.8 \cdot 10^5 \text{ m/s}^2$). Commissioning is envisaged for begin 2012.

Finally, the Bragg Institute at the Australian Nuclear Science and Technology Organization has announced that it will build a backscattering instrument to be called Emu. While the conceptual design of this instrument has not yet been fixed, commissioning is scheduled to begin in 2013.

Backscattering at spallation sources

Inverted geometry time-of-flight (TOF) spectrometers with crystal analyzer systems are a class of instruments that is widely available at pulsed neutron sources [Carlile 1992]. The basic layout for a TOF-Backscattering instrument is shown in Fig. 8. A pulsed, white neutron beam travels down a long neutron guide before striking the sample. Those scattered neutrons which satisfy the Bragg condition at the analyzer are diffracted to a multi-detector array very near the sample where they are recorded as a function of time-of-flight (TOF). As the final energy (E_s) is fixed by the analyzer, this allows the determination of the initial velocity of each of the detected neutrons and therefore the energy transfer. However instruments that routinely operate with the Bragg angle from the crystal analyzer close enough to backscattering to routinely provide resolution better than $10 \text{ } \mu\text{eV}$ have only become available over the last few years. The best example of

this trend is BASIS at the Spallation Neutron Source (SNS) at Oak Ridge, Tennessee which combines an 84 m incident beam flight path with a large area Si(111) analyzer [Herwig 2002][Mamontov 2008]

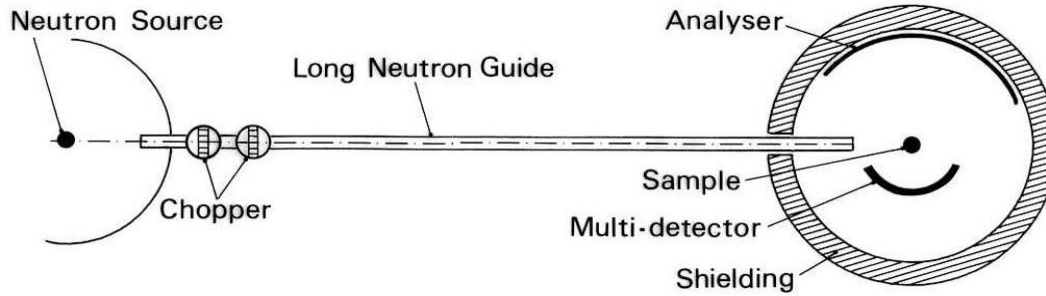


Fig. 8: Schematic diagram of a TOF-Backscattering spectrometer.

Instruments of this type tend to be more flexible than the classic reactor based instruments previously described as they provide researchers the ability to probe a wide dynamic range which can be adjusted by re-phasing the chopper system. The disadvantage is that the energy resolution is typically relaxed compared to that provided by the classic reactor-based design. For example, BASIS provides an energy resolution of 3.5 to 4.0 μeV and a typical dynamic range of $\pm 250 \mu\text{eV}$. Alternatively TOF-Backscattering spectrometers can employ mica analyzers to improve the resolution. However this approach decreases the fixed final energy, thereby decreasing the available phase space and greatly reducing the count-rate. In most cases where this compromise has been attempted, it has not proved to be scientifically productive.

An important feature of inverted geometry spectrometers compared to the more typical direct geometry TOF spectrometers described in the next section is that the available energy range in neutron energy loss is typically quite large allowing measurements to be made at very low temperatures. This comes about because direct geometry machines achieve high resolution by reducing the incident neutron energy until only a very narrow energy range is available in neutron energy loss. In addition, inverted geometry instruments do not require high-speed choppers near the sample. This allows more effective use of modern neutron optics. Thus inverted geometry spectrometers typically enjoy a somewhat higher count rate than their direct geometry counter-parts. On the other hand, due to the nearly complete freedom to choose the initial neutron energy, direct geometry instruments provide exceptional experimental flexibility.

Energy resolution for a TOF-Backscattering spectrometer

Taking the approximation that the contributions to the energy resolution (δE) from the primary TOF spectrometer (δE_p) which is given by time-of-flight and from the secondary backscattering spectrometer (δE_s) are independent and that therefore the resolutions add in quadrature (see eq. 3). For this type of instrument, the contribution to the energy resolution from the primary spectrometer is, to a good approximation, proportional to the ratio of the pulse width (Δt_p) to the total flight time t_0 :

$$\delta E_p = 2E_p(\Delta t_p/t_o) \quad \text{eq 5.}$$

Thus to improve the energy resolution, one must either reduce the width of the pulses or lengthen the instruments. If the pulse-width is reduced using a chopper, one loses intensity. Thus all TOF-Backscattering instruments that routinely operate with an energy resolution better than 10 μeV have been on long neutron guides. For example the source to sample distance on BASIS at the Spallation Neutron Source is 84 m ($t_o \approx 140,000 \mu\text{s}$) and the instrument views a poisoned moderator to limit Δt_p to 45 μs giving $\delta E_p \approx 1.3 \mu\text{eV}$.

The secondary spectrometer resolution, δE_s , is given by the same equations as those used to estimate the resolution of the classic, reactor-based backscattering instrument design (eq.(2)). Again the divergence $\Delta\theta$ of the beam when it strikes the analyzer crystals, $\delta\theta$, the average deviation (in the small angle approximation) from exact backscattering and the spread in the d-spacing of the analyzer crystals due to the Darwin width or to introduced strain are the controlling quantities. As the angular terms appear as the square of the deviation from exact backscattering, obtaining high resolution requires being very near the exact backscattering condition. For example, on BASIS the nominal value of $\delta\theta$ is 2.1° (0.036 rad) and $\Delta\theta/2$ is 0.35° (0.006 rad) for a sample that completely fills the $3 \times 3 \text{ cm}^2$ beam. Ignoring the (small) contribution from $\Delta d/d$, these two terms combine to give $\delta E_s \approx 3.6 \mu\text{eV}$. Thus the resolution of BASIS is dominated by that of the secondary spectrometer. Moreover, this example illustrates the necessity of achieving an analyzer system that operates very close to backscattering ($\delta\theta$ approaching 1°) in order to obtain an energy resolution of about 1 μeV .

An important consideration in all neutron instruments is preventing unwanted neutrons from reaching the detectors. TOF spectrometers can suffer from “frame overlap” which occurs when “fast” neutrons from one pulse catch up to “slow” neutrons in the previous pulse. This problem is particularly acute for the long flight paths needed for high-resolution instruments. To alleviate it, TOF-Backscattering instruments need complex chopper systems that limit the bandwidth and thus the dynamic range. Many factors go into the design, with the repetition rate of the source and the length of the instrument crucial. BASIS which operates at 60 Hz with a length of 84 m employs four bandwidth choppers. The number would be greater for longer instruments or for those operating at a higher repetition rate.

Crystal analyzers can also introduce spurious features through higher-order reflections which occur at energies $n^2 E_s$. Thus neutrons detected at a given time can be contaminated by a series of other energy transfers. The chopper systems can be used to eliminate all of the neutrons in the beam that have energies equal to $n^2 E_s$ for $n \neq 1$ – those neutrons which could elastically scatter from the sample and be diffracted by the higher order analyzer reflections. As elastic cross-sections are much larger than inelastic cross sections, these neutrons would cause the most serious problems. However to completely eliminate spurious scattering from the spectrum, one needs to allow only neutrons with energy E_s to reach the detector. Thus TOF-Backscattering instruments

often have a filter (typically Be) to eliminate these neutrons. Finally, the most common analyzer is Si(111) for which all of even n orders of contamination are absent due to the structure factor of Si.

The first generation of spallation source backscattering spectrometers

IRIS, operational at ISIS since 1987, is the prototypical example of the “classic” TOF-Backscattering instrument [Carlile 1991][IRIS-website] (Fig.9). In its most common mode of operation using a graphite analyzer ((002) reflection, $d=3.354 \text{ \AA}$, $\delta\theta \approx 2.5^\circ$), it provides an energy resolution of 18 \mu eV for $E_s = 1840 \text{ \mu eV}$. The instrument views a liquid hydrogen cold source via a 34 meter long curved neutron guide which terminates with a converging guide. The choppers which reduce frame overlap define the energy range which is $\pm 400 \text{ \mu eV}$ when centered on the elastic line. In addition, a Be filter (not shown), eliminates higher order reflections from the graphite analyzer.

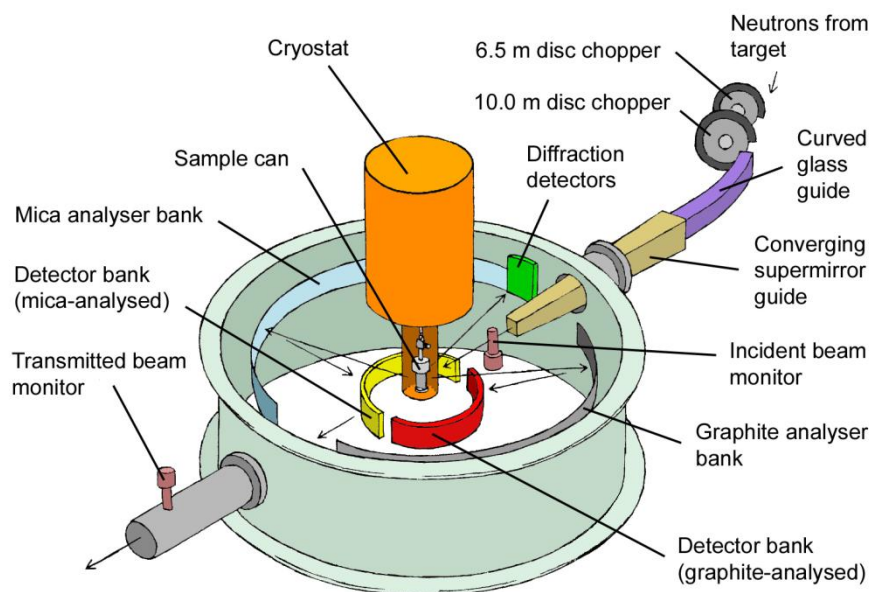


Fig.9: Spallation source backscattering spectrometer IRIS at ISIS.

The scientific necessity to attain higher energy resolution for studies of soft matter and biomolecules keeps driving quasielastic neutron scattering studies to instruments with better resolution. The easiest to attain this on an existing instrument is to effectively lengthen the instrument by decreasing the neutron energy. To accomplish this on IRIS, an analyzer consisting of an array of mica crystals which have a considerably larger d -spacing provides an energy resolution of 4 \mu eV for 9.5 \AA neutrons.

An interesting aside is that despite considerable attention to eliminating unwanted neutrons, the graphite analyzer on IRIS displayed a very high background. This was eventually traced to thermal diffuse scattering from the graphite analyzer. Cooling the entire analyzer system greatly reduced the problem [Carlile 1994]. Another issue for

graphite analyzer systems is that graphite is quite expensive. This, along with the necessity of cooling the analyzer to liquid nitrogen temperatures, limits the area subtended by the analyzer, thereby compromising the count rate. For example on IRIS's sister spectrometer, OSIRIS [Martin 1996], the analyzer covers only 8% of 4π sr compared to $\approx 20\%$ for classic reactor-based instruments [Anderson 2002].

Finally it is worth noting that the first instrument of this type was actually built at KENS in Japan. This instrument is no longer operational. [Inoue 1985]

Improving the energy resolution of TOF-backscattering instruments

The advent of the next generation spallation sources, SNS and J-PARC, has enabled a scientifically important evolution of TOF-Backscattering spectrometers to better energy resolution. The essential components of BASIS (Fig. 10) are the same as for the classic TOF-Backscattering instruments. The differences are largely in degree. First the incident beam flight path is considerably longer and the pulse width (Δt_p) shorter thereby substantially improving δE_p . Second, the analyzer is closer to backscattering which enhances δE_s . Taken together, these changes yield a substantially improved energy resolution.

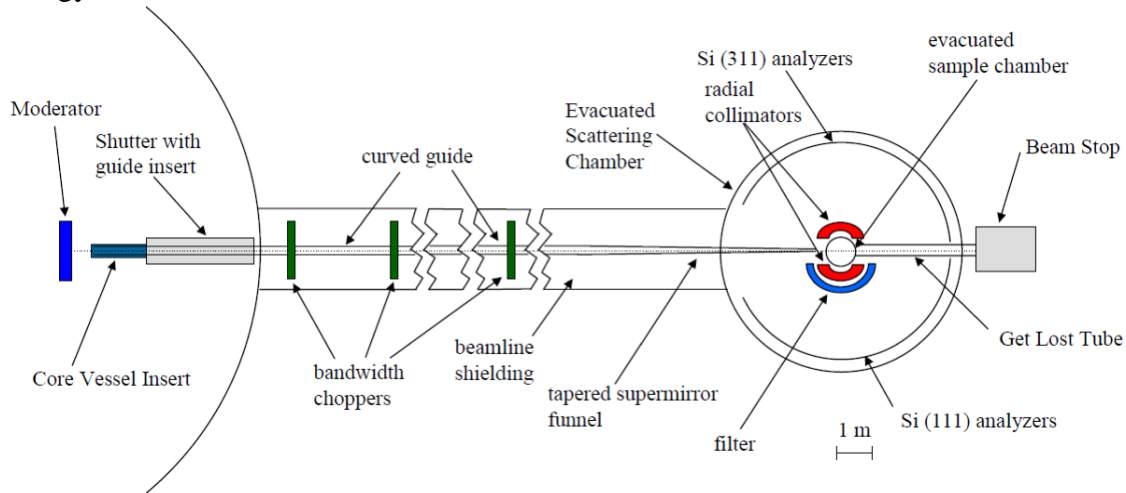


Fig. 10: Schematic diagram of the BASIS TOF-Backscattering spectrometer at Oak Ridge National Laboratory in the United States.

The secondary spectrometer provides another distinct difference. The primary analyzer is Si(111) rather than graphite (002). This has many consequences. First, Si possesses much higher energy phonons than graphite. This eliminates the need to cool the analyzer to prevent background from thermal diffuse scattering. Since Si is much cheaper, the analyzer can be significantly bigger (20% of 4π sr), enhancing the count rate.

The principle difficulty is that Si is not a mosaic crystal. Thus the crystals must be strained to enhance $\Delta d/d$ and optimize the count rate (see Eq. 3). As previously described, this is accomplished by gluing crystals of a chosen thickness r to a carefully

machined backing plate. For TOF-Backscattering instruments with $\delta\theta \approx 2^\circ$, the wafer thickness r estimated from eq.(2) should be as large as possible so that the crystals can be glued without excessive breakage. For BASIS, $r = 2$ mm.

The DNA spectrometer being built at J-PARC [Arai 2009] seeks to build on these trends. Instead of being placed on a poisoned moderator to shorten the pulse length and enhance the resolution, DNA will be placed on a coupled cold moderator with water pre-moderator. This moderator displays high peak brightness albeit with a 220 μ s pulse length. Thus DNA will employ a pulse shaping chopper to shorten Δt_p to achieve high resolution in the primary spectrometer. The energy resolution of the secondary spectrometer will also be improved compared to BASIS by placing the analyzer closer to exact backscattering (*i.e.* reducing $\delta\theta$). Due to the high brightness provided by the coupled moderator, it is expected that this instrument will provide count rates comparable to those on BASIS in spite of the resolution being ≈ 2 μ eV compared to 3.5 μ eV on BASIS. Further improvements in the resolution will likely require reducing $\delta\theta$ below $\approx 1^\circ$. This would necessitate employing a chopper to reduce the duty cycle to $<50\%$. Then neutrons diffracted from the analyzer could traverse the sample on their way to the detector while neutrons which are directly scattered from the sample to the detector would not be counted. As previously discussed, this approach is employed in all reactor-based backscattering instruments such as IN16 and SPHERES.

There is also a proposal to build a similar instrument with a Si(111) analyzer at ISIS. The instrument, called FIRES, will have an 80 m flight path and much like DNA will view a coupled hydrogen moderator and will generate short pulses using a high-speed chopper. [Demmel 2008][FIRES-website]

Optimizing TOF-Backscattering instruments for non-zero energy transfer

One problem with TOF-Backscattering instruments is that the resolution is typically matched at the elastic line. Thus as one rephrases the choppers to access large energy transfers, the energy resolution of the primary instrument is degraded compared to the that provided by the crystal analyzer. This results in a loss of intensity compared to what would be available if the two halves of the instrument were matched. The MARS spectrometer at the Paul Scherrer Institute [Treganna 2008] seeks to address this issue by placing the analyzer and detector system on movable supports so that it can be adjusted to increase $\delta\theta$ when large energy transfers are desired. Unfortunately, due to the small mosaic of the mica crystals which are employed to achieve μ eV – level resolution at the elastic line, the reflectivity of the crystals is limited as one goes away from backscattering. Thus the intensity gain is less than would be achieved if the mosaic were larger. Still this ground breaking instrument provides unique scientific opportunities when high resolution is required at energy transfers significantly away from the elastic line. And further developments to control the mosaic or $\Delta d/d$ of the analyzer crystals could yield significant improvements for high resolution measurements of dispersion-less excitations in materials.

Conclusions and outlook

The development of neutron backscattering spectroscopy, which began over 40 years ago, is in a period of rapid development. This is providing unprecedented scientific opportunities ranging from basic research to provide better understanding of physical phenomena in condensed matter to the development of future technologies such as improved materials for energy conversion and storage to better structural materials including plastics and concrete, to entirely new classes of functional nano-materials. Because many of these opportunities require understanding the dynamics over wide range of time scales, the scientific community is taking full advantage of the progress in backscattering instrumentation achieved over the last 15 years in terms of increased count rates and improved dynamic ranges. In addition, the higher count rates due to the technical advances described in this chapter may make rarely-used options like Si(311) monochromators and analyzers more attractive and may also provide the opportunity to achieve higher energy resolution using e.g. GaAs(002) crystals. Thus we eagerly look forward to the developments in the application of neutron backscattering spectroscopy in the coming years.

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