BioRef – a time-of-flight neutron reflectometer combined with in-situ infrared spectroscopy at the Helmholtz Centre Berlin

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Abstract
The novel BioRef reflectometer currently under construction in neutron guidehall-I of the BER-2 reactor at the Helmholtz Centre Berlin is designed for applications in soft matter science at solid-liquid interfaces especially under dynamic conditions. In addition to a time-of-flight (TOF) approach to be realised with three choppers in order to adapt resolutions and wavelength bands to the needs of specific applications, the instrument will be equipped with an in-situ infrared (IR) spectrometer operating in its surface sensitive ATR-geometry for simultaneous spectroscopic and conformational studies. The instrument will become operational in 2010.

1. Introduction
Neutron reflectivity measurements are a well established tool for studies of surfaces and interfaces in soft matter [1] and hard matter physics [2]. Specular reflectivity profiles provide information on the (scattering length) density profile perpendicular to the surface and hence constituted layers and layer thicknesses on a surface or interface. In contrast to monochromator based reflectometers the TOF method enables, as well as other recently proposed but not yet realised approaches [3,4], the simultaneous coverage of a defined q-range (where q denotes the scattering vector component perpendicular to the surface) without changing the angular alignment of the sample in the beam, i.e. for a fixed incidence angle \( \theta \). This makes the technique superior to monochromatic instruments in cases in which the surface structure might be time-dependent, instable or sensitive to movements. However, most importantly, the TOF approach enables to match the instrument resolution to the requirements brought forward by the system under study. This can be particularly beneficial for biological samples, where for instance ultrathin lipid membranes adsorbed to solid walls are considered.

The BioRef instrument has been designed with a strong focus on time-dependent studies under conditions like shear and flow at solid-liquid interfaces. Such dynamic conditions are important for investigations of biological model systems under more realistic physiological conditions. Corresponding sample cells and environment are developed along with the instrument taking into account the horizontal scattering geometry of BioRef. Such geometry has been chosen to allow for wide-angle scattering studies, and with respect to potential future neutron optical and spin defining and manipulating implementations in the primary and secondary reflectometer arm. The foreseen availability of simultaneous investigations of the sample by attenuated total reflection - Fourier
transform infrared spectroscopy (ATR-FTIR) with a beam geometry perpendicular to the neutron beam is especially useful for applications under dynamic conditions, i.e. changes in thickness or scattering length density of interfacial layers in an applied external field (shear or pressure) are to be linked with concurrent conformational changes of the constituting molecules.

2. Instrument design

The instrument under construction has a total length of 8.22 m and is located at the end of the 25 m long cold neutron guide NL3b in neutron guidehall-I of the BER-2 reactor at the Lise Meitner Campus of the Helmholtz Centre Berlin (former Hahn-Meitner Institute). The guide, which has a radius of curvature of 500 m, a cross section of 3 cm width and 5 cm height and which is currently coated with $^{58}$Ni provides neutrons with a minimum wavelength of approximately 3 Å and a peak flux of about $1.4 \times 10^7$ n cm$^{-2}$s$^{-1}$Å$^{-1}$ at 5.2 Å. An upgrade of the guide system with supermirror coatings has been envisaged and will change this situation significantly. It therefore had to be considered in the instrument layout. In this paper we focus on the current conditions under which the instrument will become operational. Upstream of the instrument only the perfect crystal monochromator of the V12a diffractometer [5] intersects the neutron guide with negligible influence on the neutron spectrum.

The design of the instrument is based on a three chopper system and is optimised with respect to the available space which limits the instrument length (figure 1). A first pair of choppers with variable distance defines the initial neutron pulse, while the third chopper located 3.22 m downstream of the first chopper is used to limit the utilised wavelength band. The pulse shaping choppers 1 and 2 have identical Al discs with a diameter of 0.6 m, windows of 26 deg and $^{10}$B$_4$C coating. In the range of desired wavelength resolutions $\Delta \lambda / \lambda$ from 1% to 5% they can be operated in a mode described by A. van Well [6], where the second chopper opens when the first chopper closes, i.e. with a dephasing of 26°. Consequently the wavelength resolution $\Delta \lambda / \lambda$ depends exclusively on the distance $z_0$ between the choppers (for a fixed length of the instrument) and is constant up to a certain wavelength $\lambda_0$ where the transmission has its maximum and which is defined by $z_0$, the window width D and the frequency $\omega$.

![Figure 1 Schematic view of BioRef (without shielding)](image)

The wavelength band chopper 3 with a window of 126° limits the utilized wavelength band at a maximum wavelength of $\lambda_{\text{max}} \leq \lambda_0$ and a minimum wavelength, $\lambda_{\text{min}}$, well above the cut-off of the guide system (currently at 3Å). The wavelength band chopper enables to tune the instrument resolution without changing the window size of the choppers (e.g. by double disk approaches [7]) or
the pulse frequency. All choppers are operated at the same frequency. Additionally, a variation of the chopper frequency enables a focusing of the instrument onto a certain wavelength band width and hence onto a certain q-range, which is recorded simultaneously (figure 2). This allows to optimize a specific measurement for a distinguished q-range of interest especially in cases when time resolution is required.

![Figure 2 Simulated neutron spectra at sample position as a function of chopper speed.](image)

In cases where a more relaxed resolution (\(\Delta \lambda / \lambda > 5\%\)) is required in favour of higher intensity, the approach of a constant resolution over the wavelength band can be abandoned easily (figure 3). A frame overlap mirror between the second pulse shaping and the wavelength band chopper hinders parasitic cross talk between pulses at wavelengths longer than 16 Å. Two slits define the beam collimation and hence the angular resolution. The first slit about 4 m upstream the sample position is located behind the frame overlap mirror, while the second slit, defining the footprint of the beam on the sample, is located directly in front of the sample position (distances of a few cm can be set). A third slit behind the sample limits the neutron entrance window of the shielding mounted on the secondary reflectometer arm towards the area detector of 30 cm x 30 cm. The spatial resolution is approximately 2 mm and the maximum distance between sample and detector is 2 m. A very first slit right upstream of chopper 1 is used to limit the beam width to enable high wavelength resolution and to reduce background.

The sample position as well as sample holders and substrates (Si-crystals) and a specific sample environment are being designed such that neutron reflectivity measurements can be combined with in-situ IR spectroscopy in its surface sensitive ATR-mode (Fig. 1). The last section of the neutron guide, mounted between chopper 3 and sample, is supermirror coated (m3) only on top and bottom faces like all the guide parts after the first collimation slit and is removable in order to allow for future installation of various neutron optics, polarizers or else. The whole set-up is compatible with the future upgrade of the guide system at BER-2, which includes m3 supermirror coatings at NL3b and will be discussed elsewhere.

3. Expected performance

Various simulations have been performed using the software package VITESS [8] in order to predict the performance that is expected from the novel instrument. To achieve reliable results, respectively in order to verify those, simulations were performed on the source and the guide system only. Hence, a neutron spectrum at the guide exit is provided, which has been compared to TOF measurements of the spectrum at the given position. For this purpose the latter data have been scaled by flux measurements utilizing a gold foil. Discrepancies in the flux density of more than half an order of magnitude were found. Consequently, all simulation results were downscaled accordingly. Slight differences between measurements and simulations concerning the spectral distributions have been concluded to be most
likely due to misalignments of the current guide system and do not play a major role for the results. Selected results are displayed in figure 2 and 3. In particular the expected flux values given in figure 3a are promising with respect to the design goal to achieve competitive reflectivity measurements down to a level of about $10^{-7}$ with a still reasonable q-resolution of e.g. 7% (black curve) when a background $\leq 10^{-2}$ cm$^{-2}$s$^{-1}$ is considered. The highest flux is achieved without operating the second chopper. Consequently the resolution is wavelength dependent and ranges from approximately 6 % to 14%. The angular resolution for this simulation was set to a mean value of 10%. Fig 3b shows the results achieved in the simulated measurement at a 50 nm Au layer (on a Si-substrate). The insert compares the expected resolution effects for the three cases shown in Fig. 3a.

![Figure 3](image-url)\(\text{Figure 3} \) (a) Neutron flux at detector without sample for different instrumental resolutions (left side); (b) Simulated reflectivity curve at BioRef for a 50 nm Au layer measured with 1% angular and wavelength resolution and a chopper frequency of 90Hz; the insert compares a certain q-range simulated for different resolution as shown in (a); for the [10%]-curve the chopper speed is 4600 rpm.

4. Summary
The new TOF reflectometer BioRef in its current design is dedicated to soft matter investigations at liquid-solid interfaces, especially under dynamic conditions. In addition, the set-up is highly flexible with respect to different resolution requirements as well as future upgrades and implementations with regard to a broader range of applications. The instrument will offer the unique option for complementary in-situ ATR-FTIR measurements and will become operational in early 2010.

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