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Reflectometry on curved interfaces

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ABSTRACT

Reflectometry is known since long as an interferometric method which can be used to characterize surfaces and thin films regarding their structure and, to a certain degree, composition as well. Properties like layer structures, layer thickness, density, and interface roughness can be determined by fitting the obtained reflectivity data with an appropriate model using a recursive fitting routine. However, one major drawback of the reflectometric method is its restriction to planar surfaces. In this article we demonstrate an approach to apply X-ray and neutron reflectometry to curved surfaces by means of the example of bent bare and coated glass slides. We prove the possibility to observe all features like Fresnel decay, Kiessig fringes, Bragg peaks and off-specular scattering and are able to interpret the data using common fitting software and to derive quantitative results about roughness, layer thickness and internal structure. The proposed method has become practical due to the availability of high quality 2D-detectors. It opens up the option to explore many kinds and shapes of samples, which, due to their geometry, have not been in the focus of reflectometry techniques until now.

1. Introduction

Reflectometry on surfaces and thin films is one of the oldest non-destructive techniques of interface science. Besides visible light, many other forms of waves or particles are scattered on objects and give indirectly information on the shape and structure of the object of investigation. Especially X-ray and neutron reflectometry have proven to be very powerful tools for characterization of solid state and soft matter films of nanometer up to submicrometer thickness on solid and liquid substrates [1,2]. Use of reflectometry allows to obtain detailed knowledge of structure, density, and thickness of observed layer systems and the substrate properties [2,3]. Typically films with a thickness in the range of approx. 20–2000 Å can be investigated with X-ray (XR) and neutron reflectometry (NR).

The basic prerequisite for the application of reflectrometry is a planar, reflecting sample with only moderate roughness [4], this considerably limits the general applicability of the reflectometry technique, since a large number of interesting layered samples do not fulfill this geometrical requirement, for example coated tubes, cables, shaft drive axes, or other curved or bent structures and surfaces.

At this point it has to be emphasized, that there are already curved objects known and used for reflecting X-ray or neutron radiation, e.g. as optical elements or beam guides [5]. However, these optics have different intention and mode of action, than the reflectometry method suggested in this paper. A short overview shall highlight these differences.

X-ray radiation and also neutrons can be effectively guided on curved interfaces as far as the impinging angle is smaller than the critical angle, or for multilayered beam guides (e.g. *supermirrors* [6]), as their cut-off angle, respectively. In 1948 Kirkpatrick and Baez introduced a mirror system of two elliptically curved focussing mirrors for imaging objects with X-rays [7]. This method was further developed to systems with variable curvature and focus [8,9]. However, already since the twenties of the last century, it is known, that cylindrical capillaries and tubes are able to guide radiation internally [10,11], the transport of radiation remains possible, when the capillary is slightly curved.

In 1948 Kreger used tapered glass capillaries to focus X-rays at small samples (0.1 mm³) for diffraction experiments [12]. Balaic et al. improved this capillary optics by using capillaries with a

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paraboloidally reducing diameter in 1995 [13] which increased the focal length and made them applicable for X-ray microscopy.

Also (cold) neutrons have a perceptible critical edge towards low-refractive index materials, thus, optics similar to those for X-rays are imaginable. Neutrons can be guided with curved mirrors [5,14,15], and can be concentrated with dedicated mirror systems in a focal point, these optics are called *focussing benders* [16] . For X-ray and neutron radiation poly-capillary optics exist, which are used for focussing the beam sections similar as with a lens [17–19]. A unique extra option for neutron beam modification provide so-called *polarizing benders*. These specially curved neutron guides can polarize the neutron beam, since the critical angle of the impinging neutrons differs for their different polarization [20].

All these mentioned optical elements have in common, that they either guide the entire beam, or map source points to focal points, to increase local intensities and spacial resolutions. This functionality is especially needed for scanning and transmission microscopic and spectroscopic purposes, or for diffraction of very small structures or objects. The optics combine high reflectivity and extremely low roughness. The working mechanism of all these optics bases on total external reflection at grazing incidence. For a comprehensive review of these advanced X-ray and neutron optics see for instance Ref. [21].

In comparison the method of *reflectometry at curved interfaces* is different and has additional requirements and challenges, which shall be addressed here shortly. Reflectometry is using multiple specular reflections of coherent radiation at layered structures [4]. Here the reflected waves interfere with a locked phase shift (or path difference). The resulting scattered intensity as a function of the incoming angle exhibits characteristic interference pattern (maxima and minima), which finally allows characterising the films thickness, refractive index, and roughness.

Out of these considerations, the following requirements for using reflectometry ensue:

1. A relateable geometry of reflection is a vital prerequisite, i.e. assignable angle correlation and parallelism of layers.
2. Defined phase correlation between the reflected wave portions must exist, which means again, parallel layers or interfaces and, additionally, an adequately coherent radiation.
3. The final requirement is detectability, i.e. sufficient reflectance at the buried interfaces and suitable detector geometry and efficiency to sense the interference pattern. In case of curved objects, this list of demands extends by 3 additional requirements:
4. The reflected intensity is spread over a larger solid angle, thus the fraction of coherent radiation is reduced.
5. Two extra parameters have to be aligned (see Fig. 1. *vertical (normal)* and *parallel rotational (rolling)* axis).
6. Finally, waviness of substrates or films can cause undefined angular correlation and has to be taken in consideration.

This paper proofs, that the alignment of a curved sample is practicable, that the divergence of the reflected beam remains low enough. Coherence and general reflectance is sufficiently high, and, most important, the phase relation between the portions of reflected beam remains locked.

Despite of the mentioned difficulties, there have been already a few studies done in which X-rays were applied to curved samples. Lesslauer et al. and also Rapp et al. have used an experimental set-up where the reflection plane and the axis of curvature of the sample were orthogonal to each other [22–25]. In this special geometry the sample is irradiated simultaneously under a range of different angles of incidence. The observed reflectivity curves showed Kiessig fringes representing the interference pattern corresponding to the total film thickness and, additionally, Bragg peaks due to ordered lipid double layers which were the subunits of these multilayer films. This diffractive method is primarily applicable for layered (quasi-crystalline) films, overall film thickness and lamellar spacings can be determined, together with the electron density of the subunits. However, films with irregularly varying scattering density, which do not exhibit Bragg peaks, are less suited for this technique. Due to the simultaneous appearance of various impinging angles, non-specular scattering superimposes

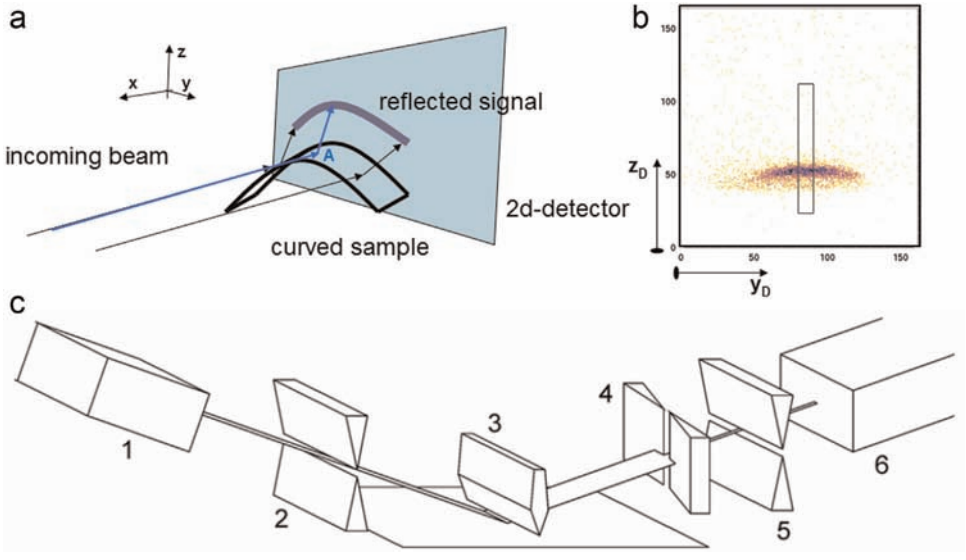

Fig. 1. Scheme of reflectometry (a) on a curved interface, where the axis of curvature lies within the instrumental reflection plane as defined in the main text. Only the beam reflected from the summit line of the sample may be interpreted in the same manner as in the case of a planar sample. (b) Detector image obtained at N-REX+ during the measurement of a curved sample. The black rectangle symbolizes the software mask applied to prevent the merge of the laterally off-set intensity with the centrally reflected intensity signal. These off-set portions originate from reflection on non-vertically oriented sample surface and thus correspond to deviating angles of incidence, not fully identical to the instrumental angle setting θ. (c) Experimental set-up: positions of the X-ray source (1), horizontal slit of the X-ray source (2), sample slit to reduce direct beam intensity (knife slit) (3), additional vertical slit (4), the horizontal slit of the detector (5) and the detector (6). Shown on example of a planar sample.