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ADAM, the new reflectometer at the ILL

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Abstract

The new reflectometer ADAM at the ILL is described and some of the results obtained in the first year of operation are presented. These include a reflectivity of a Si wafer over eight orders of magnitude, a measurement of a thick [^{56}Fe/^{57}Fe] isotope superlattice, and a polarised reflectivity of a Co/Cu multilayer. The instrument is now available to outside users.

Keywords: Neutron reflectometry; Polarisation analysis; Instrumentation

Throughout the last decade layered materials of unprecedented structural quality have become available. Many fascinating new properties have been found in thin-film systems including polymers, biological membranes, and magnetic multilayers and superlattices. Neutron reflectometry (NR) was developed to make use of the well-known advantages of the neutron as a probe for the study of such thin-film systems [1]. Currently NR is one of the fastest growing neutron techniques.

The new reflectometer ADAM (Advanced Diffractometer for the Analysis of Materials) at the ILL in Grenoble/France is one of the latest additions to the fleet. After a design and construction phase of about one year at the Ruhr-Universitaet Bochum, the instrument was moved to the ILL in February 1996. In May 1996 ADAM saw its first neutrons. Since then, the machine has been systematically tested, first with an unpolarised beam, and since September 1996 with a polarised beam and polarisation analysis.

ADAM is situated on ILL's neutron guide H 53 which is fed by a liquid deuterium cold source. In Fig. 1 a schematic sketch of the instrument is shown. Making use of a focusing HOPG monochromator a fixed wavelength of $\lambda_0 = 4.4$ Å is selected. A Be filter removes any higher harmonics λ_0/n, $n = 2, 3, \ldots$ which are also reflected by the monochromator. Optionally, the Be filter can be removed, allowing the use of $\lambda_0/2$ [2] to extend the scattering vector range. Two motorised collimation slits, positioned 2 m apart, provide the high collimation required for reflectometry experiments. Being a fixed wavelength instrument, specular reflectivity measurements are carried out by standard $\theta/2\theta$ scans, whereas off-specular measurements can be performed, e.g. by θ-rocks. The scattering plane

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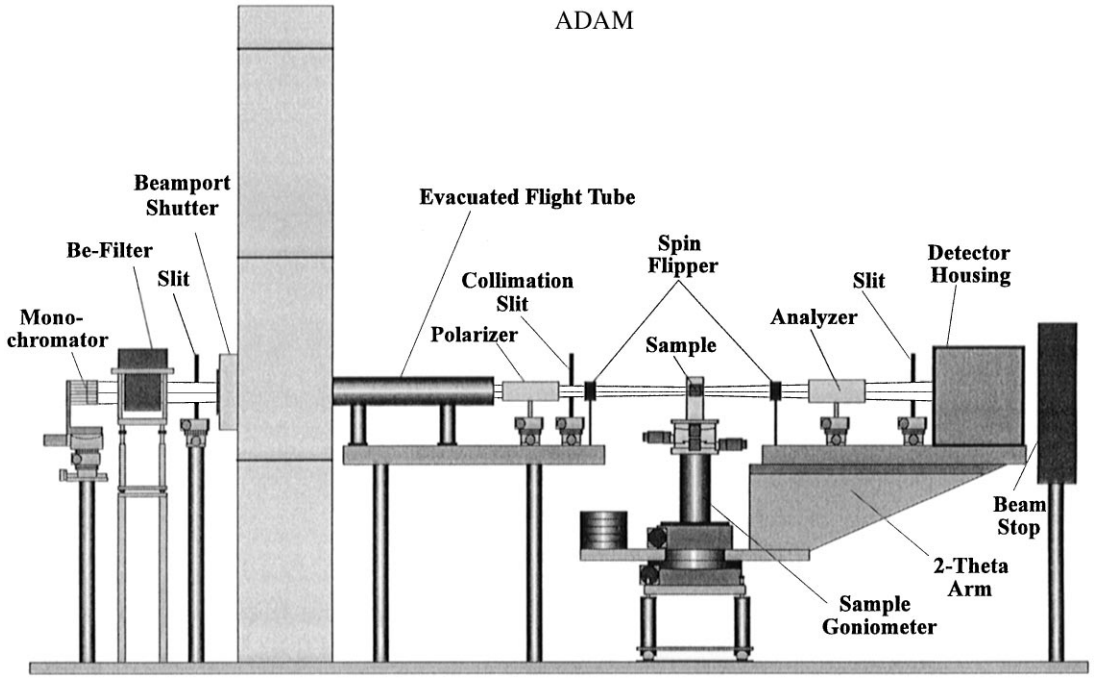

Fig. 1. Schematic side view of the ADAM reflectometer at the ILL in Grenoble/France.

is horizontal, i.e. the samples are mounted vertically on a motorised heavy load goniometer head which provides all motions for the alignment of the sample.

As detailed e.g. in Refs. [3,4] polarised neutron reflectometry (PNR) is ideally suited for the study of magnetic thin-film systems. For this purpose, optional supermirror polarisers and flippers in front of and behind the sample are available. The use of transmission polarisers [5] obtained from the HMI in Berlin/Germany avoids the need to realign the diffractometer in the polarised mode. Furthermore, it will be possible to detect two polarisation cross-sections at the same time by simultaneously measuring the transmitted and the reflected beam of the analysing supermirror behind the sample. For this purpose two ^{3}He pencil detectors are mounted in the well-shielded detector housing. One of these detectors is motorised to allow easy alignment with the beam which is reflected by the analysing supermirror. The ability to measure two cross-sections at the same time can reduce the measuring times by a factor of two. A two-dimensional position-sensitive detector will become available in 1998 significantly reducing counting times for off-specular measurements.

As sample environment a customised electromagnet and cryofurnace combination is available. For measurements on large samples a sample holder with an integrated heater up to 150°C can be provided. Instrument and sample environment are controlled by the commercial SPEC diffractometer control software running under LINUX on a Pentium PC.

Although primarily designed for small-angle reflectometry, ADAM can access 2θ angles up to about 140°. Thus, scattering vectors $\boldsymbol{Q}$ up to 5.4 Å^{-1} can be reached when using $\lambda_0/2$ (i.e. without the Be filter). This allows the investigation of ordering phenomena on atomic scales (via Bragg scattering) at high Q and on the nanometer scale (via reflectometry measurements) at small Q in one experiment and within the same sample environment. For example, such information has been

of crucial importance for the understanding of the correlation between the Cr antiferromagnetic order and the coupling between the Fe layers in Fe/Cr superlattices [6].

To demonstrate the instrument's dynamic range we show a reflectivity measurement of a Si wafer of 5″ diameter in Fig. 2. To minimise background, it was essential to put the sample into a vacuum chamber eliminating air scattering. The remaining off-specular intensity was measured independently and subtracted. The resulting specular reflectivity essentially corresponds to a Fresnel reflectivity with a broad bump superimposed which can be ascribed to the native oxide of the Si. Clearly, reflectivities up to more than 0.5 $Å^{-1}$ and down into the 10^{-8} range can already be achieved with ADAM.

In the first year of operation at the ILL measurements from magnetic, isotope, and polymer multilayers have already been performed. Whereas the latter data are discussed elsewhere in these proceedings [9] we will present examples of the other experiments to highlight the capabilities of the instrument.

In Fig. 3 data taken from an epitaxial $^{57}Fe/^{56}Fe$ superlattice of nominal composition Pd 50 Å/^{56}Fe 90 Å[^{57}Fe 10 Å/^{56}Fe 90 Å]$_{15}$/MgO (substrate size 4 cm^2) grown by sputtering are shown. The sample had been obtained from the University of Uppsala/Sweden. Ranging over nearly six orders of magnitude, a Q range up to 0.4 $Å^{-1}$ and six superlattice peaks were obtained. Although the expected nominal overall thickness D_{nom} of the sample is quite large, Kiessig fringes rapidly oscillating at $\Delta Q_z = 2\pi/D_{exp}$ are easily resolved up to 0.11 $Å^{-1}$ (see insets). The overall thickness $D_{exp} = 1653$ Å determined from the Kiessig fringes agrees well with the nominal value. Good agreement is also found between the superlattice period Λ determined from the separation of the superlattice reflections and the nominal superlattice period. However, upon close inspection of the higher-order reflections a second component is observed, indicating a partial deviation from the nominal superlattice period. A more detailed and quantitative account on this work will be given elsewhere. For the present purpose, we would like to point out that such a structural analysis can only be carried out

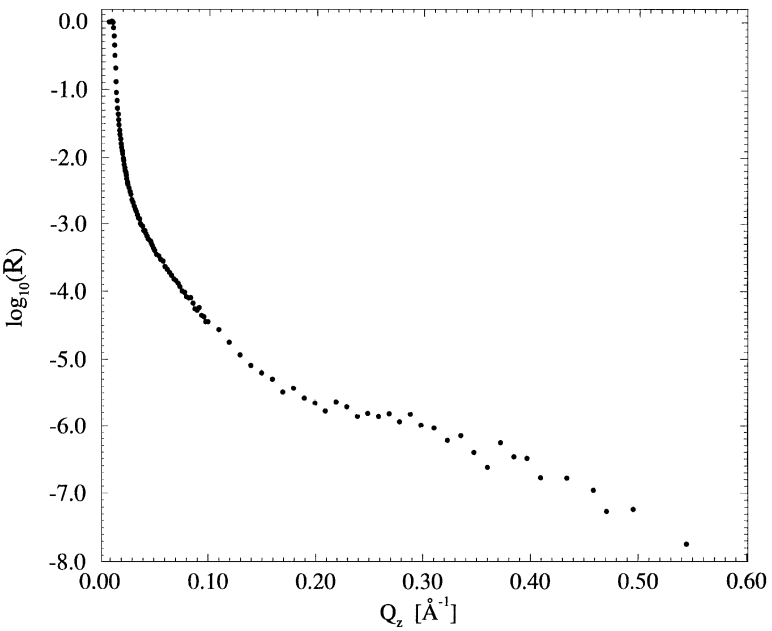

Fig. 2. Reflectivity of a Si wafer measured over nearly eight orders of magnitude with ADAM. For details see text.

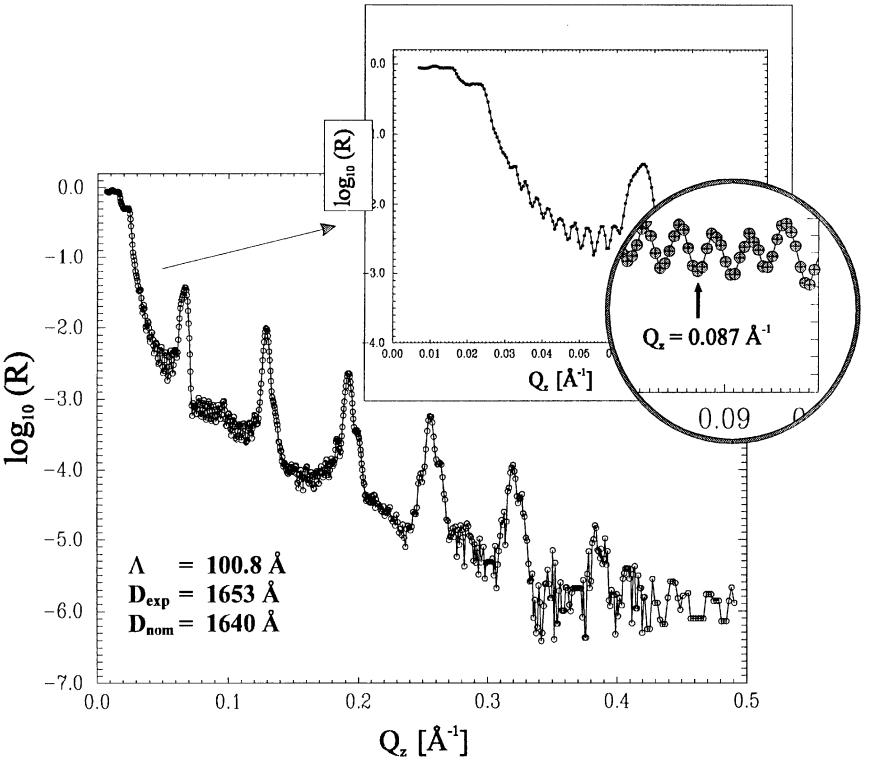

Fig. 3. Reflectivity of an epitaxially sputtered superlattice of nominal composition Pd 50 Å/^{56}Fe 90 Å[^{57}Fe 10 Å/^{56}Fe 90 Å]$_{15}$/MgO (substrate size 4 cm^2). In the insets blowups of the measured reflectivity are shown making the very short period Kiessig fringes visible. The line is a guide to the eye.

with neutrons, since no scattering contrast between isotopes for the same element exists for X-rays. Furthermore, this example demonstrates the high Q-resolution and intensity of ADAM, easily allowing to resolve Kiessig fringes from thick samples. Clearly, much thicker samples could be investigated which might be interesting, e.g., for industrial coatings.

In our final example we want to demonstrate ADAMs capability for polarised measurements. In Fig. 4 we show two reflectivities measured with polarisation analysis on a sputtered [Cu 15 Å/Co 34 Å]$_7$/Al_2O_3 multilayer in the two different schematically depicted orientations. The sample has ferromagnetically aligned in-plane magnetisations in the Co layers. Furthermore, it exhibits the useful feature of a strong uniaxial anisotropy along an easy axis marked 'EA' in the right-hand side of the figure. Thus, by reorienting the sample in a small guide field the sensitivity of PNR to the orientation of the in-plane magnetisation can be studied. This orientational sensitivity is depicted schematically in the right part of Fig. 4. The axis marked NSF (non-spin flip) is parallel to the polarisation direction ***P*** of the incident neutrons. It indicates that the spin state of the incident neutrons is not flipped by a magnetisation oriented parallel to this axis. The axis marked SF (spin flip), on the other hand, indicates spin flip of the neutrons for any magnetisation components along this axis. Thus, by separating NSF and SF scattering via polarisation analysis, PNR is sensitive to the orientation of any in-plane magnetisation. A more detailed account of the underlying theory can be found e.g. in Refs. [3,4]. The orientational sensitivity of PNR has also lead to the term "vector magnetometry" for the technique.

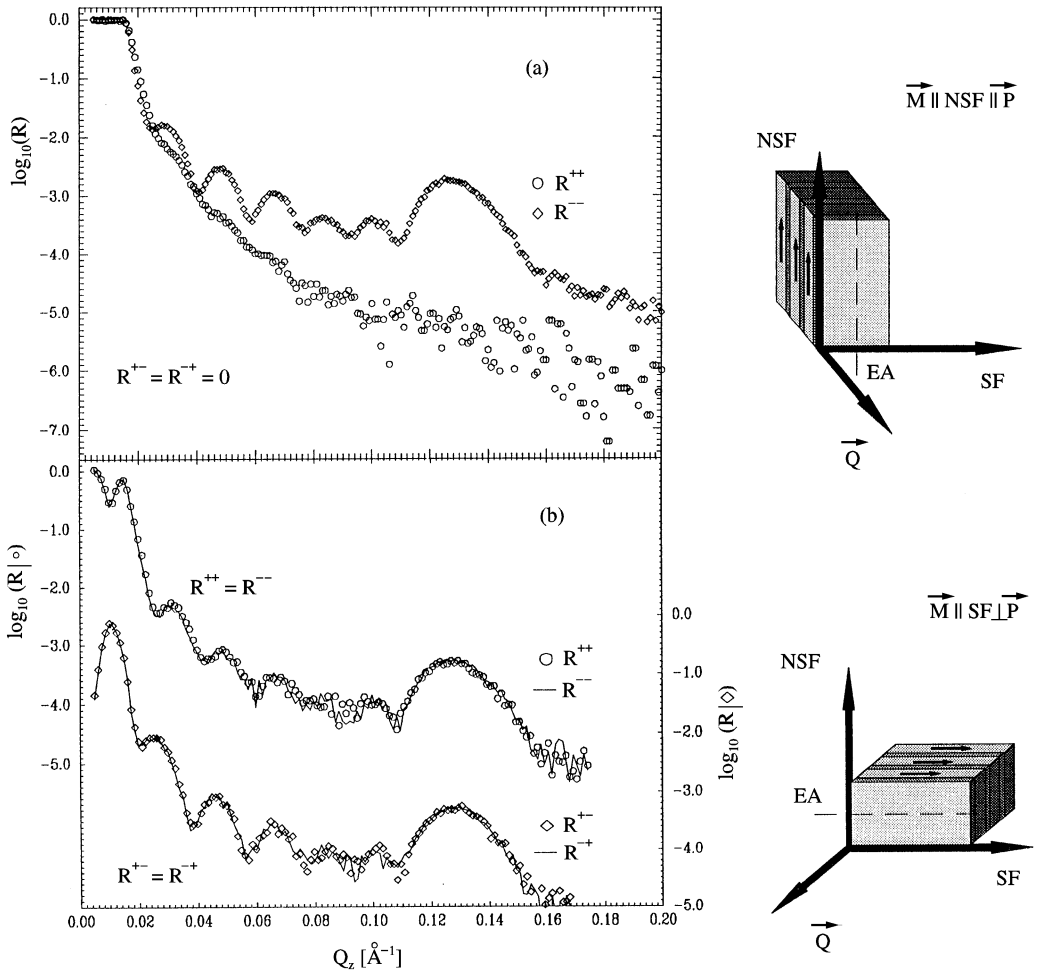

Fig. 4. Reflectivity of a [Cu 15 Å/Co 34 Å]$_7$/Al_2O_3 multilayer measured on ADAM using the polarisation analysis option. On the right-hand side the scattering geometry is depicted with the scattering vector $\boldsymbol{Q}$ perpendicular to the film plane and the NSF (non-spin flip) and SF (spin flip) axes, as described in the text. (a) and (b) show the measured reflectivities for the two sample orientations shown on the right.

A recent study of collinear and non-collinear coupling in exchange-coupled superlattices with this technique can be found in Refs. [7,8].

The data in Fig. 4a were taken with the sample oriented with the magnetisation parallel to the NSF axis. Consequently, no SF scattering is observed. The splitting of the two NSF reflectivities R^{++} and R^{--} is a measure for the magnitude of the magnetisation of the sample. Here, the superscripts indicate the polarisation state in front of and behind the sample, respectively. In Fig. 4b, on the other hand, the sample is oriented to cause maximum SF scattering. Consistent with theory, strong degenerate R^{+-} and R^{-+} reflectivities are found. Furthermore, the splitting of the NSF reflectivities has completely vanished since the magnetisation component along the NSF axis is zero. The observed NSF reflectivity now results from purely nuclear scattering. The data exhibit a superlattice peak due to the periodic structure of the sample and five Kiessig fringes due to the sample's finite thickness. Only in R^{--} in Fig. 4a hardly any

structure is observed in the data. This is due to a very similar scattering length density of the Co, the Cu and the substrate for this case. A more thorough discussion, including quantitative fits to equivalent data from the same sample taken on another reflectometer can be found in Ref. [3].

For the present purpose, we want to demonstrate ADAMs capability to perform such measurements with high quality. Notably, at least five orders of magnitude in reflectivity are accessible although the sample has a surface of only 2 cm^2. The measured splitting of nearly three orders of magnitude between the two NSF reflectivities at the superlattice peak around 0.13 $Å^{-1}$ indicates a good efficiency of the polarisation elements. Flipping ratios up to 54 are achieved, i.e. the polarisation efficiency is larger than 98%.

In conclusion, we present a new high flux reflectometer to an international user community. Two modes of access are in effect. First, 30% of the ADAM beam time is made available via the standard ILL proposal scheme. Second, German users can obtain beam time via the 'Verbundforschung'. For details see http://www.ep4.ruhr-uni-bochum.de/adam.html.

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