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CRISP, A PULSED SOURCE NEUTRON REFLECTOMETER FOR SURFACE STUDIES

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A time-of-flight neutron reflectometer, installed on the spallation neutron source, ISIS, is described (1).

Particular features of the time of flight method: fixed sample geometry and sample illumination, constant resolution in $\sin\theta/\lambda$, and simultaneous measurement over a wide range of $\sin\theta/\lambda$ for approximately constant statistical accuracy, are highlighted.

Examples of its potential use to study problems in surface chemistry and surface magnetism are presented.

Preliminary commissioning results are included.

Introduction

In recent years a number of neutron optical phenomena have been investigated (2). Critical reflection of neutrons has been extensively exploited in neutron scattering measurements and techniques, especially in the applications of neutron guide tubes (3,4), and neutron spin polarisers (5,6).

More recently neutron reflectivity measurements above the critical reflection angle have been applied to investigate interfacial phenomena (7-9) and surface magnetism (10,11).

Traditionally critical reflection measurements using cold neutrons have been made on reactor sources by performing θ-2θ scans with a well monochromated and collimated beam. The wavevector transfer resolution in such measurements is determined by both $\delta\theta$ and $\delta\lambda$. For fixed collimation $\delta\theta$ and monochromatisation $\delta\lambda$ the resolution is a function of the angle of incidence θ through a $\cot\theta\delta\theta$ term. Furthermore the area of the sample illuminated varies with θ necessitating geometric corrections.

More recently time of flight techniques have been exploited by Felcher (10,11) on a pulsed neutron source, and by Farnoux (12) on a reactor source. There are three main advantages in using a white beam time of flight technique on a pulsed neutron source like ISIS:

(i) The reflectivity profile is measured in a fixed geometry. The range of $\sin\theta/\lambda$ measured is determined by a wide band of incident wavelengths, $\Delta\lambda$, at a fixed angle of incidence, θ. The area of sample illuminated is constant.

(ii) There are also consequences for the resolution in $\sin\theta/\lambda$. On ISIS this resolution is determined only by the collimation, $\delta\theta$, as the time of flight resolution contribution from the $\delta\lambda$ term is $\delta t/t \sim 0.5\%$, which is negligible compared with typical values from the divergence ($\cot\theta\delta\theta$) term ($\sim 5\%$). As a result the resolution is essentially constant over the $\sin\theta/\lambda$ range measured.

(iii) Reflectivity and spectral profiles combine to give a slowly varying measured intensity over most of the $\sin\theta/\lambda$ range of the measurement. It is therefore possible to collect data with approximately the same statistical error at all $\sin\theta/\lambda$ values in a fixed measurement time.

Instrument Design and Specification

A general schematic plan of the instrument is shown in Figure 1. The need to investigate liquid surfaces dictates that the incident beam should be inclined to the horizontal at an appropriate reflection angle. The wavelength range needed in the measurements must extend as far as possible into the cold neutron region and the instrument is therefore located on the N4 beamline, viewing the 20K H_2 moderator.

A horizontal slit geometry with an incident beam ~ 50 mm wide and ~ 2 to 4 mm high is used and is collimated at a fixed inclined angle of 1.50° to the horizontal plane. This is the highest inclination angle that can be constructed conveniently in the ISIS shutter apertures; it allows a sufficiently large $\sin\theta/\lambda$ for the liquid surface studies - 0.052 Å^{-1} at λ = 0.5 Å. The sample position is located at 10.25 m from the source and the sample-detector distance is 1.75 m at which single He_3 gas detector is located.

A single disc chopper, located at 6 metres from the moderator is used to define the wavelength band $\Delta\lambda$ and to partially suppress frame overlap contamination. Additional suppression of long wavelength frame overlap neutrons is provided by a series of thin Ni films evaporated onto single crystal silicon mirrors and which are inclined to the incident beam at the critical reflection angle corresponding to the shortest wavelength which can produce frame contamination.

Horizontal slits after the mirrors and before the sample position define the beam collimation and size.

An option to insert a supermirror polariser, magnetic guide fields and a spin flipper between the normal sample position and the frame overlap mirrors has been included to enable polarised neutron experiments to be performed.

A wavelength range, $0.5 \leqslant \lambda \leqslant 26.4$Å gives a range in $\sin\theta/\lambda$ of $.001 \leqslant \sin\theta/\lambda \leqslant .052$Å^{-1} at a fixed angle of incidence $\theta = 1.5°$. This represents the maximum $\sin\theta/\lambda$ range available for experiments on liquid surfaces. For solid surfaces this range can be extended to higher and lower values of $\sin\theta/\lambda$ since the angle of incidence can be changed by rotating the sample out of the horizontal plane. Many of the polarised beam experiments on surface magnetism will for example be performed on solid surfaces at much smaller incident angles, typically $\theta \sim 0.5°$.

The resolution in $\sin\theta/\lambda$ has two contributions: a $\cot\theta\delta\theta$ term and $\delta\lambda/\lambda = \delta t/t$ term. $\delta t/t$ is given by a convolution of the moderator pulse width, δt_{mod}, and the time channel width in the time of flight electronics. δt_{mod} is ~ 100 µs for energies $\leqslant$ 10 meV and ~ 20 λ for energies $\geqslant$ 10 meV, thus $\Delta t/t \sim 5 \times 10^{-3}$ for a 12 m flight path. If the time of flight channel width is ~ 100 μsec then the resultant $\delta t/t$ is ~ 0.7%. The resolution required in $\sin\theta/\lambda$ is ~ 2 to 5% and therefore on the ISIS is determined essentially by the contribution from $\cot\theta\delta\theta$. The collimation angles required to achieve resolutions ~ 2 to 5% in $\sin\theta/\lambda$ are ~ 1 mrad. For solid surface experiments at reflection angles $\theta \leqslant 1.5°$, such resolutions are easily maintained by reducing the slit widths and hence divergence $\delta\theta$.

Science

Critical reflection of neutrons provides information on the scattering density or refractive index variations perpendicular to a surface. We will highlight particularly the application to problems in surface chemistry and surface magnetism.

The use of contrast variation, through deuterium-hydrogen exchange, is particularly important for hydrogenous system, and is therefore relevant to a number of problems in surface chemistry. Calculations for a number of such systems have been made (7), and some measurements are also available (8,9). Typical systems that have been considered include Langmuir-Blodgett multilayers, soap films, density profiles at a liquid-vapour interface, insoluble monolayers at a liquid surface, adsorption of soluble organic materials at a liquid-vapour interface, wetting phenomena, adsorption at the surface of an electrode, and polymer film surfaces and interfaces.

The potential of the technique is illustrated with a calculated example of a polymer film (9a). Figure 2 shows the reflectivity profile for a 200 Å $pmma_{d8}$ film on a P_sh_8 substrate, where the solid and dashed lines correspond to a sharp and diffuse boundary.

Magnetic materials have a spin dependent refractive index, will give rise to two different reflectivity profiles R^+ and R^-. This has been used extensively as the basis for the production of polarised neutrons by critical reflection. Critical reflection of polarised neutrons is a particularly sensitive probe for problems in surface magnetism. Felcher has published (10,11,13) results and calculations of the critical reflection of polarised neutrons applied to a range of problems in surface magnetism. These include the measurement of the magnetic flux penetration into a superconducting film of niobium (11), calculations of the contribution to the reflectivity profile of the magnetic dead layer on a ferromagnetic surface (10), and of surface critical phenomena in ferromagnets (13).

A particularly fascinating current area of research is that of magnetism at surfaces and interfaces. Freeman et al (14) have reviewed the status of the theoretical modelling of surface magnetism and has drawn attention to several discrepancies between reported results. Although early models showed that the surface layers of a ferromagnetic like Ni were magnetically dead, later calculations by Wang and Freeman (15) and Jepsen et al (16) showed that the Ni(001) surface has a moment similar to that in the bulk. In contrast to these results Wang and Freeman's calculations on Fe(001) (17) showed a large increase in the surface magnetism for a 7 atomic layer calculation; similar results have recently been reported by Victoria et al (18).

For the purposes of illustration we show the flipping ratios (R^+/R^-) calculated for the magnetisation profile published by Victoria et al (18) for Fe(100) in Figure 3; the reflection angle used was $\theta = 1.5°$. Enhanced magnetism effects only become apparent at the shorter wavelengths (~ 5Å) or larger ($\sin\theta/\lambda$), where the reflectivities are $R^+ \sim 10^{-3}$, $R^- \sim 10^{-4}$.

Preliminary Commissioning

Some initial commissioning of the CRISP spectrometer have now been carried out. The initial aim has been to obtain a reflectivity profile and hence optimise the spectrometer geometry and reduce background levels.

Initial operation has been without the frame overlap chopper: as a result the wavelength band is limited to 0.5 to 6Å, and to beam sizes $\leqslant 1.5$ mm.

Shown in Figure 4 as the solid line is the measured reflectivity profile for a 750Å copper film on a glass substrate; also plotted as the dashed line is the calculated reflectivity.

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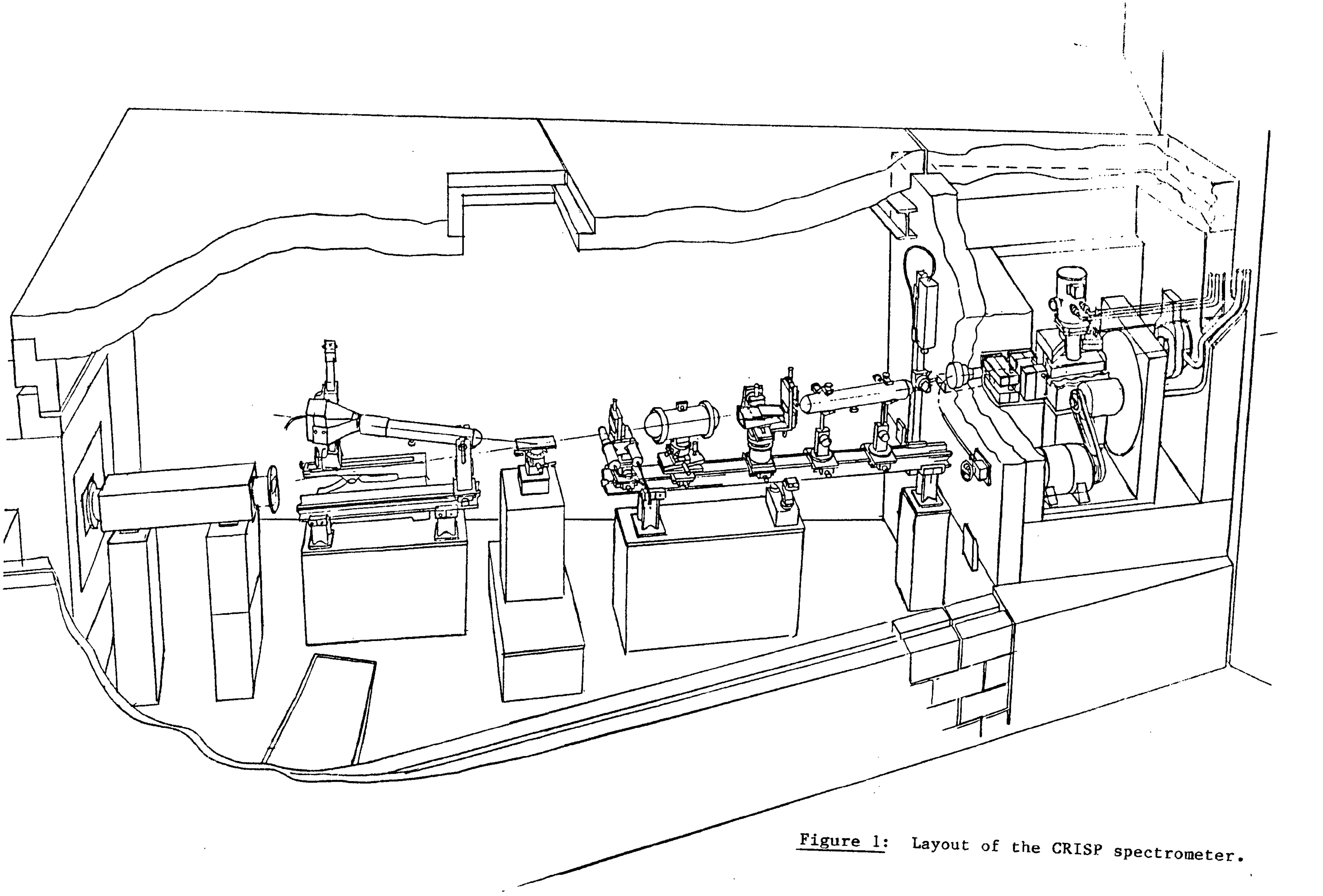
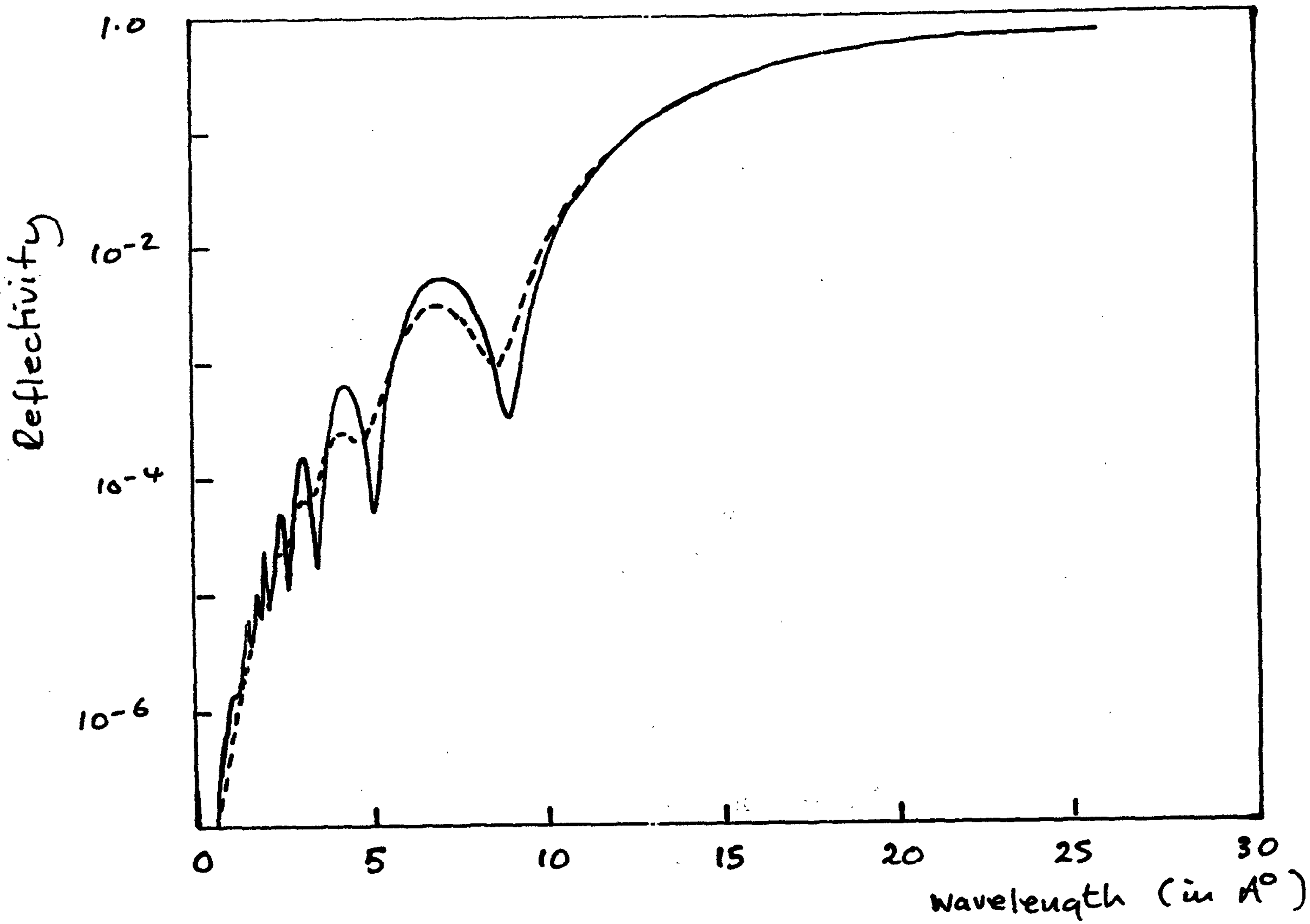


Figure 1: Layout of the CRISP spectrometer.

Figure 2: Calculated reflectivity profile for 200Å $pmma_{d8}$ film on a P_sh_8 substrate with (i) —— sharp and (ii) --- diffuse boundary.



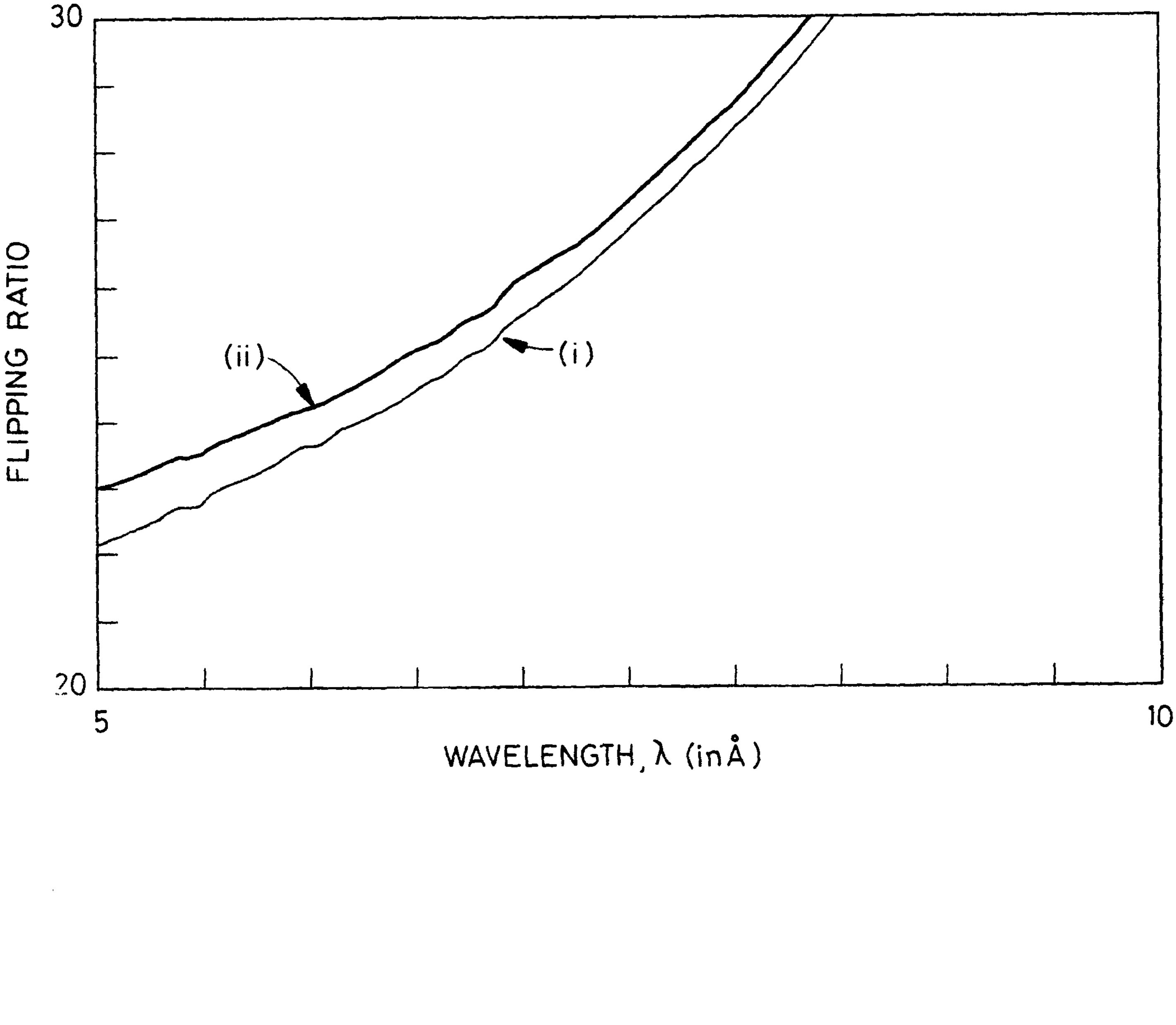
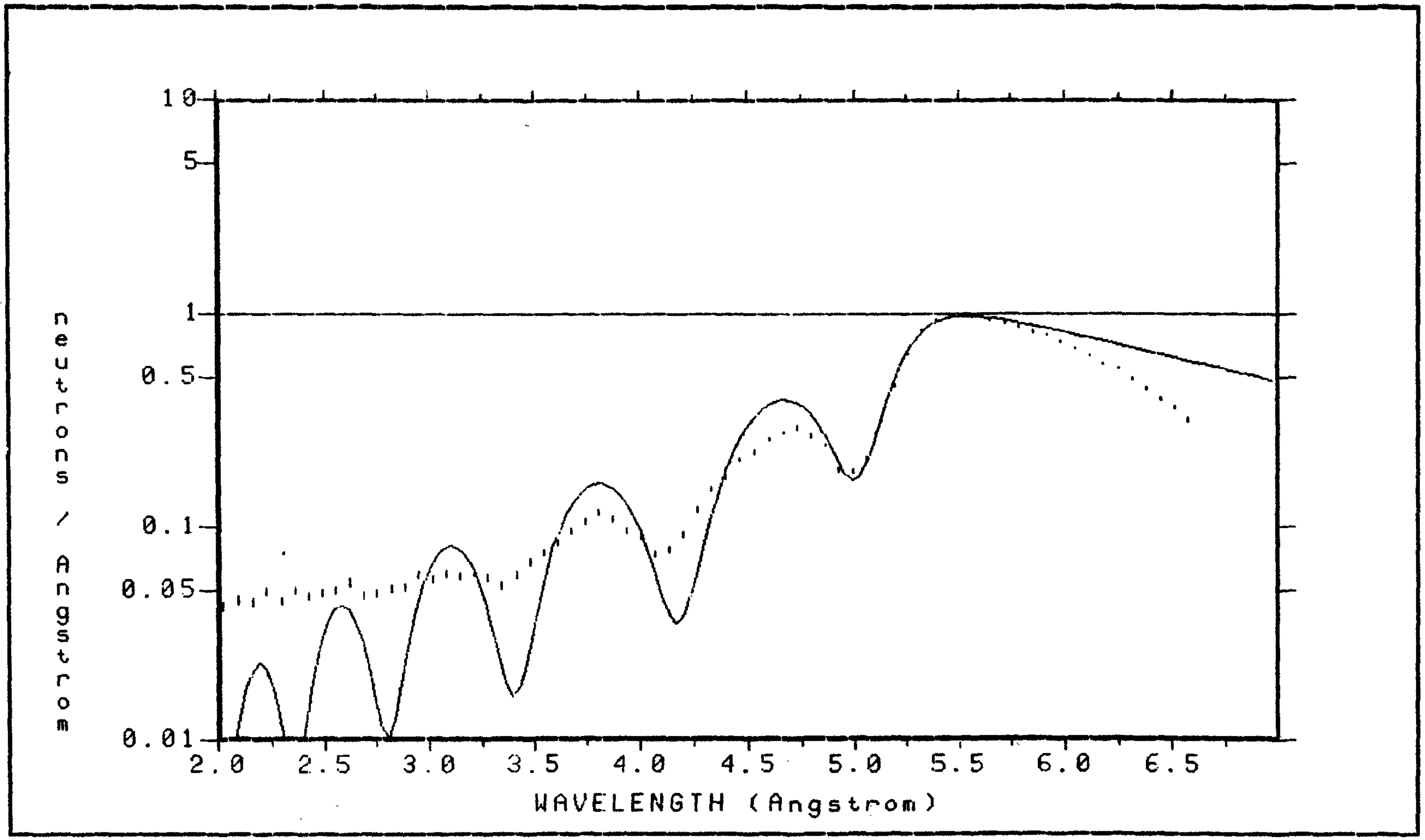


Figure 3: Flipping ratio (R^+/R^-) versus wavelength for (i) bulk iron, (ii) magnetisation profile published by Victoria et al (18) for Fe(100), $\theta = \theta = 1.5°$, $\delta\theta = 0.075°$.



<u>Figure 4</u>: Reflectivity profile for 750 Å copper film in glass substrate (i) ——— measured on CRISP, ~ 10 μAhrs, θ = 1.5°, Δθ ~ 5%, and (ii) --- calculated reflectivity profile.