Cryst. Res. Technol.	**37**	2002	7	645–653

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Grazing Incidence In-plane Diffraction Measurement of In-plane Mosaic with Microfocus X-ray Tubes

Dedicated to Prof. Hildebrandt on the occasion of his 80th birthday.

The in-plane mosaic structure of Au/Fe and GaN-based epitaxial layers has been determined directly by laboratory-based grazing incidence in-plane x-ray diffraction in which Bragg reflections normal to the plane of the wafer are probed. High intensity and acceptable signal-to-noise can be obtained with no modifications to commercially available equipment. Excellent agreement is obtained between measurements of the same Au/Fe multilayer samples at the European Synchrotron Radiation Facility in Grenoble and with the laboratory system employing a focused x-ray beam from a microfocus generator. The technique is particularly important for the GaN-based systems as it uniquely provides a measure of the so-called twist mosaic independent of the out-of plane (tilt) mosaic.

Keywords: diffraction, x-ray

(Received March 21, 2002; Accepted May 1, 2002)

Introduction

Grazing incidence in-plane x-ray diffraction[1] (GIIXD) is an x-ray scattering technique through which the in-plane lattice parameter and lattice orientation of very thin surface and buried semiconductor layers can be determined. With the incident beam close to or below the critical angle for total external reflection, a Bragg reflection is excited from planes perpendicular to the surface (fig. 1). As the depth penetration of a wave incident below the critical angle is only a few nanometres, the technique has become of major importance in studying the surface crystallography and, in particular, the reconstruction of semiconductors. Pioneering experiments were undertaken *in-situ* in ultra-high vacuum during molecular beam epitaxy[2] but there has been increasing application for the study of quantum dot and wire structures *ex-situ*[3,4,5]. Variation of the grazing incidence angle permits the in-plane lattice parameter to be probed as a function of depth[6].

Unlike bulk diffraction, the scattering is intrinsically weak, the geometry being such that only the surface truncation rod is cut rather than the scattering vector passing through the reciprocal lattice point. When combined with the loss of intensity through beam spill-off at these very low angles and the need to restrict the beam size in the sample plane in order to achieve reasonable resolution, it is understandable why almost all experiments have been

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conducted at synchrotron sources. Only a very few university laboratories have undertaken in-house experiments. The combination of the difficulty of performing in-house measurements or the need to travel to use infrequent periods of synchrotron radiation beamtime has meant that GIIXD has not been adopted by industrial scientists working in the semiconductor field.

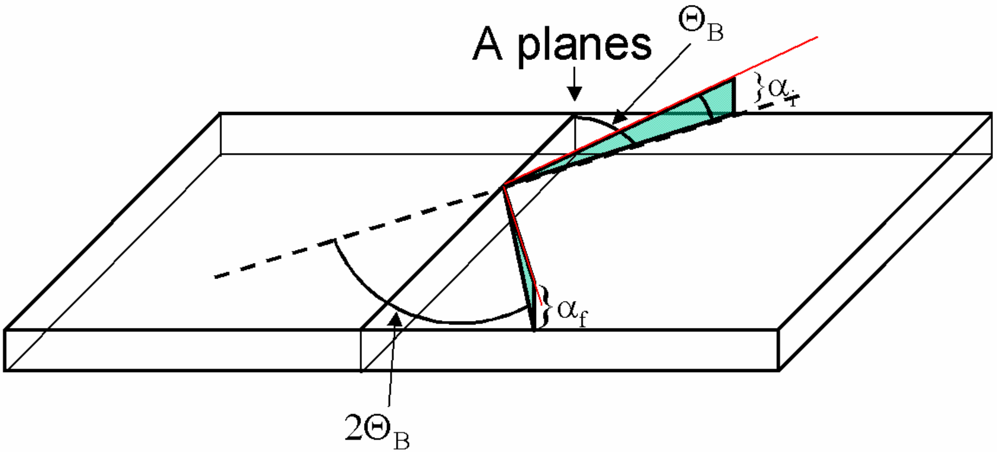

Fig. 1: Grazing incidence in-plane x-ray diffraction (GIIXD) geometry showing Bragg reflection from the planes marked A.

Recently, however, major improvements have occurred in the brilliance of laboratory x-ray sources due to the application of novel x-ray optics[7]. With either total reflection ellipsoidal mirrors[8] or polycapillary devices[9,10], beams of typically 0.25 mm diameter and 2-3 mrad divergence can be delivered at the sample from a microfocus x-ray generator closely coupled to the x-ray optic. Such beam size and diameter are well matched to the optimal requirements for GIIXD[11]. In this paper we report the successful application of a laboratory-based microfocus x-ray source with focusing optics to the direct measurement of in-plane mosaic in epitaxial Fe/Au multilayers and GaN based epilayers. The metal multilayer results are compared directly with equivalent data taken on beamline BM28 at the ESRF in Grenoble.

Experimental Requirements

Experiments have been performed using a Bede D1 diffractometer fitted with a Microsource® microfocus x-ray generator for high-resolution thin film powder diffraction in addition to a standard sealed source and Bragg reflection optics for high-resolution diffraction. No modification to the commercial system was undertaken, although ease of use could have been improved by raising the detector slightly out of the horizontal plane. The Cu target x-ray generator was run at a power of 80W, the beam being focused onto the sample using a Reflex Micromirror™ ellipsoidal mirror of focal length 300 mm and divergence at the sample of 2 mrad in both horizontal and vertical directions. The spot diameter was 0.3 mm full width at half height maximum (FWHM). No further conditioning of the incidence beam was used. The sample was rotated in the 200mm XY stage of the diffractometer until it was horizontal, the vertical height being adjusted to half-cut the incident beam. A small rotation about the χ axis, now horizontal and perpendicular to the incident beam, permitted the grazing angle to be tuned. The sample was rotated about the Φ axis, normal to the sample surface. A 2 mrad divergence Soller slit limited the angular acceptance of the scintillation detector in the horizontal plane in which the detector was scanned (2Θ axis). Over the small range of grazing incidence angles used in GIIXD, the vertical aperture of the detector was large enough to accept all beams. While it is straightforward to modify the apparatus to scan a detector of limited vertical aperture in the vertical direction, all measurements reported here were for the open detector, thereby integrating the scatter down the surface truncation rod.

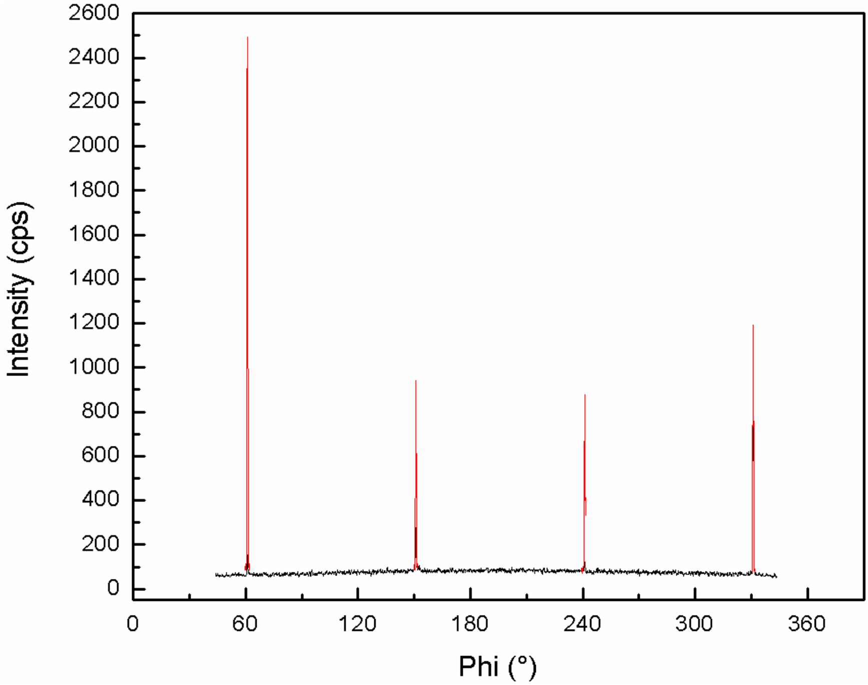

Fig. 2: 220 grazing incidence diffraction peaks from a silicon wafer, optimised for the maximum intensity at each point.

Results

The instrumental resolution was determined by recording the grazing incidence diffraction peaks from a (001) orientation, 6 inch diameter silicon wafer. The detector was set for the 220 reflection from the CuKα lines and the sample scanned about the Φ axis. Four peaks were detected (fig 2) separated by 90°. The size of the specimen was such that these could not arise from bulk scattering from the edge of the wafer and represent genuine surface diffraction peaks. As a function of the grazing incidence angle χ, all peaks showed a maximum at about the critical angle. However, the peak heights were by no means equal and the precision of the Φ rotation of the commercial stage was insufficient to compare intensities in the four peaks, even when there was no variation due to differing spill off from an irregular shaped sample. Figure 2 shows a composite of the four reflections, each adjusted for the maximum intensity on a χ scan. The angular variation in the background, though barely visible in fig 2, also arises from the wobble on the Φ axis.

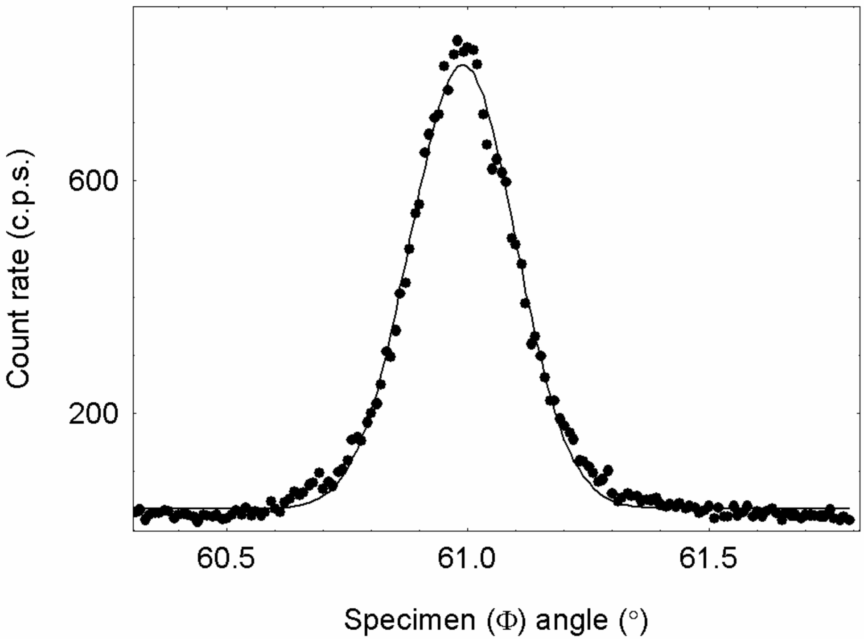

Fig. 3: Width of the 220 peak on a Φ scan of the silicon wafer.

Figure 3 shows an expanded view of one of the peaks and the instrumental resolution of the Φ scan that measures the sample mosaic. We obtained a value of 0.26 ± 0.005° for the Φ FWHM, a resolution entirely consistent with the Soller slit aperture and incident beam divergence in the horizontal plane. A Φ-2Θ scan through one of the 220 peaks is shown in figure 4. The FWHM of the scan, measured from the detector motion was 0.17 ± 0.01°.

The peak intensity is found to be a sensitive function of the grazing incidence angle, despite the open detector integrating along the surface truncation rod. Between 0.1 and 0.2° the rise in intensity is almost linear (fig.5), corresponding to the sinusoidal change in the beam footprint. Beyond the critical angle, the intensity falls as the beam penetrates in to the sample and the reflectivity falls. The reduction in background signal as the incidence angle is increased is a result of partial shielding of the detector from the main beam scatter and fluorescence, the χ axis not coinciding with the diffraction vector direction.

A direct comparison between synchrotron radiation and laboratory GIIXD data was performed on a series of epitaxial Au/Fe multilayers grown by molecular beam epitaxy at the University of Leeds. GIIXD measurements performed on the multi-axis XMaS diffractometer on BM28 at the ESRF played a key part in the recent identification of electron channelling in (001) oriented Au/Fe multilayers grown on MgO[12]. Compared with (111) orientation Au/Fe multilayers grown on the a plane of sapphire, the current-in–plane saturation conductivity was larger for the (001) multilayers of equal individual layer thickness. Similarly, for constant Fe thickness, the gradient of the saturation conductivity as a function of Au thickness was greater for the (001) than for the (111) multilayers. The greater conductivity of the (001) orientation multilayers was determined not to be from lower interface scatter, as grazing incidence diffuse scatter showed that the (001) multilayer interfaces were three times rougher than those of the (111) multilayers. High-resolution x-ray diffraction[13] confirmed the relative interface roughness and showed that the out-of-plane mosaic was greater for the (001) multilayers[14].

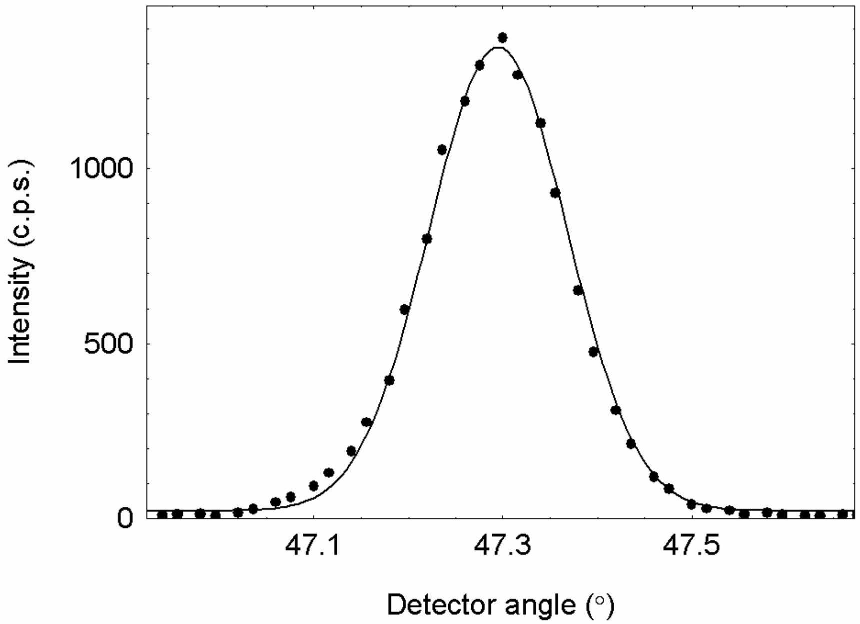

Fig. 4: Φ-2Θ scan through one of the Si 220 peaks.

Figure 6 shows the in-plane mosaic measured by GIIXD in the laboratory and on beamline BM28 at the ESRF. The sample had 20 repeats of (Au 12Å Fe 10Å), there being in addition a 20Å Au seed layer. In all cases, the FWHM of the Φ scan is very substantially greater than the instrumental resolution. The curves can be fitted well to Guassian functions. For the (001) Au/Fe multilayers, excellent agreement is found between the FWHM measured at the ESRF (3.92 ± 0.005°) and in the laboratory (3.78 ± 0.005°). In both situations, the contribution from the instrumental resolution function is negligible. As an x-ray wavelength of 1Å was used at the ESRF compared with the 1.54Å CuKα radiation in the laboratory, we conclude that the FWHM represents almost entirely the in-plane mosaic rather than a microstructural length scale.

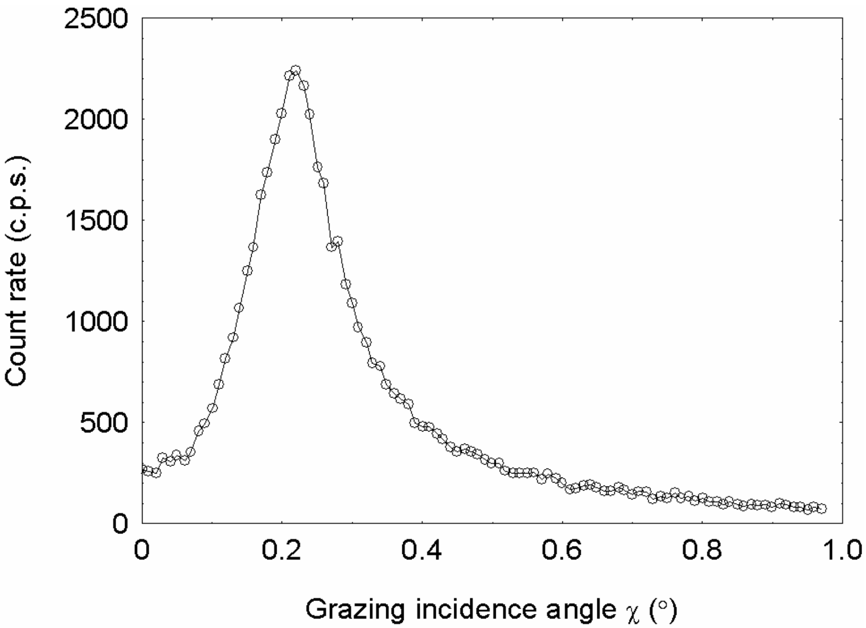

Fig. 5: Intensity as a function of the grazing incidence angle χ for the Si 220 reflection

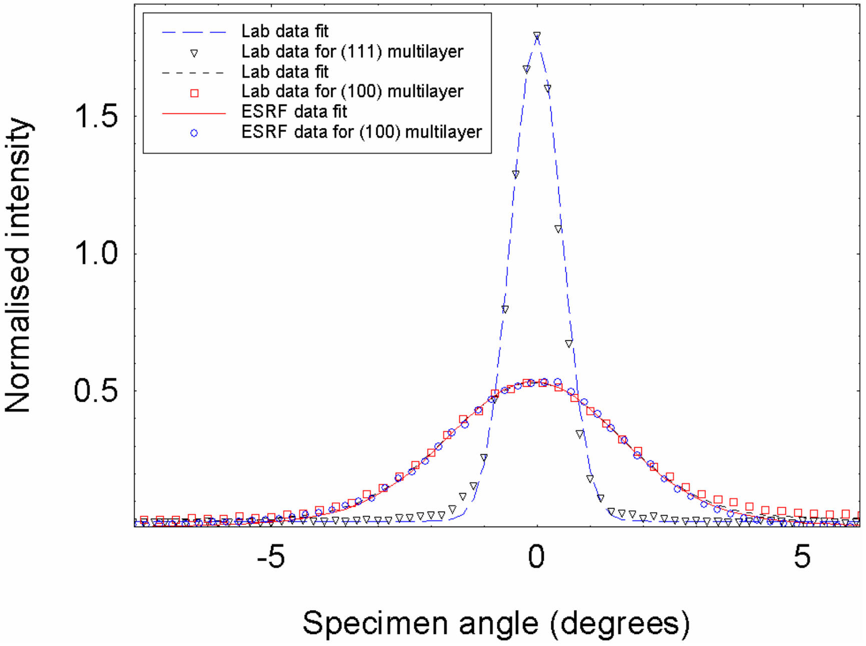

Fig. 6: In-plane mosaic widths for the two orientations of epitaxial Au/Fe multilayers recorded in the laboratory with a focused beam and at the ESRF. The points represent (unsmoothed) experimental data and the lines are Gaussian fits.

For the (111) orientation Au/Fe multilayers, the agreement is poorer. The FWHM measured in the laboratory was 1.02 ± 0.01° compared with the 0.80 ± 0.005° measured previously at the ESRF. This discrepancy may be due to degradation of the multilayer due to oxidation; the time between the ESRF and laboratory measurements was over a year.

Measurement of the mosaic structure of GaN and related ternary compounds is of major importance in the control of growth of short wavelength visible optoelectronic devices. The large mismatch between substrate and epilayer results in very high misfit dislocation densities and associated high threading dislocation densities in sub-grain boundaries in the epilayers. Since the first triple axis x-ray diffraction studies on GaN[15], it has been found that the rocking curve broadening observed in double axis measurements is almost all due to mosaic tilts, there being very little strain present.

Determination of the out-of-plane mosaic, or tilt distribution, of the epilayer is very straightforward, as this comes directly from the rocking curve width of a surface symmetric high-resolution x-ray diffraction measurement. Much more difficult is the determination of the in-plane mosaic, the so-called twist distribution. This involves measurement of a series of skew symmetric reflections[16] and a complex analysis[17] that is of dubious reliability. GIIXD provides a much more direct method for the determination of the in-plane mosaic of GaN-based compounds.

A Φ scan through the truncation rod of one of the 11.0 peaks of a GaN/GaAlN multilayer is shown in Figure 7. The peak fits well to a Gaussian function of FWHM 0.32 ± 0.005° Assuming that the widths add in quadrature, we determine an in-plane, or twist mosaic, of 0.19± 0.01°. In agreement with other reports, this is large in comparison with the tilt mosaic distribution of 0.022 ± 0.001° determined from the surface symmetric 00.2 reflection in a high-resolution diffraction scan.

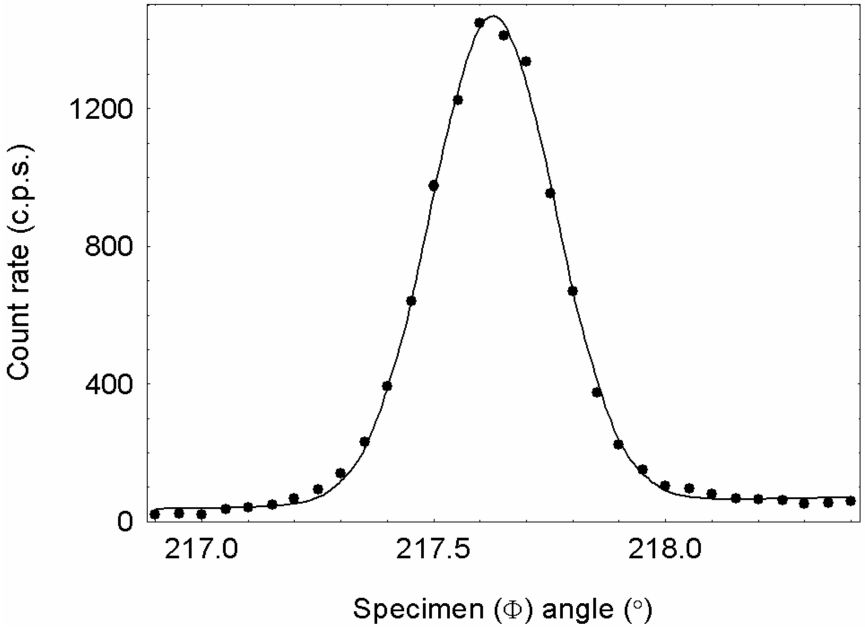

Fig. 7: Φ scan through the 11.0 peak of a GaN/GaAlN multilayer showing the in-plane mosaic width convolved with the instrument function. The point are experimental data, the line is a Gaussian fit.

Discussion and Conclusions

Prior to this study, almost all GIIXD measurements were undertaken at synchrotron radiation facilities, as the signal to noise in laboratory experiments was extremely poor. Only a few laboratories have persisted in making measurements with conventional sources. MacDonald's group at Cardiff use a curved monochromator with a standard sealed source[18] and they can detect peaks from epitaxial layers as thin as 1 nm. Kobayashi *et al*[19] used a Rigaku diffractometer with a channel cut Ge monochromator and a slit to determine, as here, the twist mosaic in GaN directly from the GIIXD rocking curve width. (Their twist value for organo-metallic vapour phase deposited GaN was very comparable to that reported in this paper.) The count rate is not given in the paper of Kobayashi *et al.* but the Poisson noise on the data indicates that the count rates were very low. (The out-of-plane and in-plane data in Fig. 1 of their paper appear to have been inadvertently switched.) However, use of a focused beam from a microfocus generator provides orders of magnitude gain in flux and makes these experiments extremely easy to undertake.

While use of an energy dispersive detector improves significantly the GIIXD signal to noise ratio through the rejection of fluorescence, the background with a standard scintillation detector proves acceptable for the study of nanometre thickness films. A further improvement can be achieved with a Ni filter to reduce the Bremstrahlung and CuKβ line. However, to achieve the signal levels reported here, further monochromatisation is not permissible and a weighted average of the Kα lines must be used for in-plane lattice parameter determination.

As indicated, the GIIXD technique not only provides direct determination of the in-plane mosaic, but can also be used to determine the in-plane lattice parameter as a function of depth from the surface. In such measurements on Si/Ge on Si we have used the apparatus described here with a polycapillary optic. Here, an optic of low (2 mrad) divergence was used. Such a polycapillary bundle delivers a much larger diameter beam (5 mm) at the sample than the ellipsoidal mirror, but for large wafers this is an advantage. Further, use of a Soller slit as the analyser means that the width of the beam normal to the incidence direction does not degrade the resolution. The intensity in the GIIXD peaks can be between one and two orders of magnitude greater than with the ellipsoidal mirror.

By surface science standards, the structures investigated here are extremely thick, but the high intensity observed here indicates that structural studies of extremely thin layers on semiconductor surfaces may be viable using focused beams from microfocus generators. For the study of in-plane mosaic and relaxation in films tens of nanometres thick, the GIIXD technique is very straightforward to implement.

Acknowledgements

Thanks are expressed to Dr P A Ryan, Dr D E Joyce and Mr J Wall of Bede plc for assistance in setting up the x-ray optical elements. The financial support of EPSRC and the technical help from the XMaS beamline staff at the ESRF in collecting the synchrotron radiation data is gratefully acknowledged. MSG thanks the University of California Trust (UK) for a travel award and gratefully acknowledges a UC-SMART grant. BKT thanks the Leverhulme Trust for the award of a research fellowship.

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