J. Synchrotron Rad. (1998). **5**, 226–231

Focusing Optics for High-Energy X-ray Diffraction

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(*Received 4 August 1997; accepted 20 January 1998*)

Novel focusing optical devices have been developed for synchrotron radiation in the energy range 40–100 keV. Firstly, a narrow-band-pass focusing energy-tuneable fixed-exit monochromator was constructed by combining meridionally bent Laue and Bragg crystals. Dispersion compensation was applied to retain the high momentum resolution despite the beam divergence caused by the focusing. Next, microfocusing was achieved by a bent multilayer arranged behind the crystal monochromator and alternatively by a bent Laue crystal. A 1.2 μm-high line focus was obtained at 90 keV. The properties of the different set-ups are described and potential applications are discussed. First experiments were performed, investigating with high spatial resolution the residual strain gradients in layered polycrystalline materials. The results underline that focused high-energy synchrotron radiation can provide unique information on the mesoscopic scale to the materials scientist, complementary to existing techniques based on conventional X-ray sources, neutron scattering or electron microscopy.

Keywords: focusing optics; high-energy synchrotron diffraction; strain gradients.

1. Introduction

The interaction of high-energy X-rays with matter is weak compared with interactions in the classical crystallographic X-ray energy range. This makes high-energy X-ray diffraction a unique tool if sufficient flux can be provided. For example, the diffraction from single crystals is not biased by extinction and the bulk properties can be studied in complicated environments due to the low absorption. When structural polycrystalline materials are studied, many fundamental quantities (grains, composites, cracks, strain fields) have dimensions on the micrometre scale and a diffraction probe with a micrometre spatial resolution is highly desirable (Poulsen *et al.*, 1997). At the ESRF, a dedicated experimental station is being developed aiming to provide a three-dimensional diffraction probe volume of $5 \times 5 \times 50$ μm to investigate the bulk properties of polycrystalline materials (Kvick & Poulsen, 1997). The experiments employing one-dimensional microfocusing described here are pre-studies within this project.

High-energy third-generation synchrotron sources combine, for the first time, high-energy X-rays with a narrow source size, opening the possibility for microfocusing. Focusing optics are essential as they provide several orders of magnitude more flux compared with the use of narrow apertures. In the following, first the status of the development of a focusing energy-tuneable fixed-exit monochromator based on a combination of bent Laue and Bragg crystals is reported. The bent Laue–Bragg monochromator scheme was suggested recently (Suortti & Schulze, 1995) and the commissioning has now reached the stage at which first experiments are performed. Two novel developments will enlarge the range of applications of this narrow-band-pass multipurpose monochromator. It is demonstrated that microfocusing perpendicular to the

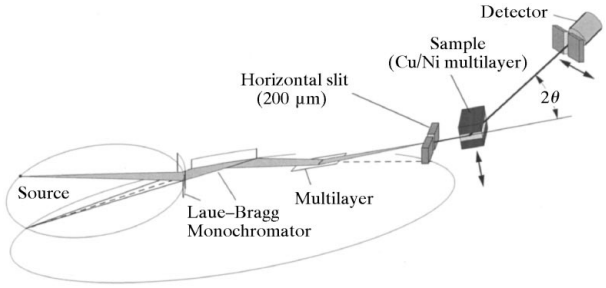

Figure 1
Top view of the Laue–Bragg monochromator and multilayer combination. The experimental set-up on BM5 is sketched. The horizontal focusing of the bending-magnet radiation is achieved by meridionally bent Laue and Bragg crystals. The vertical focusing is provided by a bent multilayer. Residual strain profiles were measured by translating the sample through the line-focused beam monitoring the scattering angle. The parameters of the set-up are compiled in Table 1.

Table 1
Parameters of the Laue–Bragg monochromator multilayer combination as tested on BM5.

The particular choice of Laue and Bragg crystals may be used between 40 and 100 keV. The flux was measured using an Si PIN diode.

Energy	68.5 keV
Laue–Bragg monochromator	
Source–monochromator distance	31.5 m
Monochromator–focus distance	8.5 m
Reflection	[511]
Thickness of Laue crystal	1 mm
Energy bandwidth, $\Delta E/E$	4×10^{-4}
Efficiency	40%
Accepted beam divergence	1 mrad
Horizontal focus size (FWHM)	500 μm
Multilayer	
d-spacing	25 Å
Illuminated length	220 mm
Focal distance	0.75 m
Lateral d-spacing gradient	13%/200 mm
Average bending radius	415 m
Aperture at 68.5 keV	800 μm
Vertical focus size (FWHM)	4 μm
Peak reflectivity	75%
Rocking-curve width, $\Delta\theta/\theta$	1.4%
Slope error of substrate (FWHM)	2.5 μrad
Flux behind multilayer	10^{10} photons s^{-1} $(0.1\ A)^{-1}$

scattering plane can be achieved by a combination with a curved multilayer. It is also shown that the Laue–Bragg geometry allows one to overcome, in a practical way, a resolution broadening due to the focusing divergence by dispersion compensation. Secondly, the microfocusing capabilities of curved Laue crystals are demonstrated. Both optics provide microfocusing only in one dimension and were applied to measure the residual strain gradients in layered polycrystalline materials, such as coatings and structural multilayers.

These composite materials are developed to improve specific properties like hardness, corrosion and wear resistance (Nix, 1989). The local residual stress is a fundamental quantity in the analysis and modelling of materials properties. The diffraction techniques probing the residual stress in materials use the lattice plane spacing as strain gauge, utilizing Bragg's law which relates the lattice plane spacing to the scattering angle. We present a novel technique which allows a direct measurement of residual-strain depth profiles. The potential of the technique is demonstrated on a Cu/Ni multilayer structure but a complete characterization of the strain and stress states is beyond the scope of this paper.

2. Laue–Bragg monochromator

The Laue–Bragg monochromator consists of a combination of meridionally bent Laue and Bragg crystals as sketched in Fig. 1. It provides a focusing energy-tuneable fixed-exit monochromator in an energy range ~30–140 keV. The low-energy limit is set by the absorption in the Laue crystal, whereas the efficiency decreases at higher energies with the kinematical diffraction limit of the required higher-order reflections.

A crucial advantage compared with sagittal focusing, the standard monochromator scheme up to 30 keV, arises from the insensitivity to anticlastic bending, which now takes place perpendicular to the scattering plane. Furthermore, the meridional bending increases the integrated reflectivity by typically one order of magnitude giving an energy bandwidth a few times wider than for a perfect Si [111] crystal, although higher-order reflections can be used to increase the Bragg angles. If better resolution is required, the bandwidth broadening might be avoided by using a symmetric Laue crystal (Cole & Brock, 1959). In this case, the intrinsic energy resolution of the employed high-order reflection may be obtained as there is no broadening due to the beam divergence as in the sagittal geometry. The parameters of the Laue–Bragg monochromator shown in Fig. 1 are listed in Table 1.

2.1. *Status of commissioning*

Laue–Bragg monochromator prototypes were developed and tested on ESRF wiggler (ID15) and bending-magnet (BM5) beamlines. Bending mechanisms allowing for crystal cooling were developed which allow dynamical bending and therefore energy scans of the monochromator. A horizontal beam fan of up to 60 mm width (2 mrad) was accepted at BM5. At ID15, the energy was tuned from 87 keV to 120 keV by dynamically adjusting the bending radii of the crystals. The prototype monochromators were operated in air and a settle time of 10 min was required when the full wiggler beam impinged onto the monochromator. The power in the incident beam after the attenuators is estimated as 180 W from which 38 W should be deposited in the water-cooled Laue crystal. A horizontal focus size of 500 μm (FWHM) was obtained. This broadening is not due to figure errors of the curved crystals but is due to the crystal thicknesses and the polychromatic divergences upon asymmetric reflections. The Rowland circle geometry and the requirement of fixed-exit geometry do not allow this limitation of the focus size to be overcome.

2.2. *Dispersion compensation*

An inherent problem in high-energy diffraction arises from the small Bragg angles which lead to a large broadening of the momentum resolution by a beam divergence as described by the differentiated Bragg equation

$$\Delta d/d = \Delta\theta/\tan\theta. \quad (1)$$

At typical Bragg angles of 0.1 rad, a focusing divergence of 1 mrad limits the resolution to $\Delta d/d = 1\%$. Although the d-spacing, taken as the peak centre, can still be extracted to a relative accuracy of 10^{-4}–10^{-5}, the information contained in the peak shape will be hidden by the wide instrumental function. This limitation can be overcome for powder-like samples using a slit in front of the detector as

a resolution-defining element, if the beam is focused on the detector slit. However, the diffracting volume in the sample is increased at the same time. In a different approach we reduced the resolution broadening by applying an energy gradient over the focusing divergence such that a parallel beam exits from the sample (Fig. 2). This dispersion compensation allows an improvement of both the spatial and the momentum resolution. The required energy gradient was produced by changing the bending radii of the crystals. The matching condition is given by

$$2\tan\theta_S = \tan\theta_{\rm LB}/|1 - \rho_{\rm mon}/\rho|, \qquad (2)$$

where θ_S and $\theta_{\rm LB}$ are the sample and monochromator Bragg angles and ρ and $\rho_{\rm mon}$ are the actual and monochromatic (Rowland circle geometry) bending radii, respectively. θ_S and $\theta_{\rm LB}$ are typically comparable such that dispersion compensation would be achieved by increasing or decreasing the bending radii from the Rowland circle diameters by factors of 2 or 2/3, respectively. The dispersion matching strictly applies only for one sample reflection and it is therefore of practical importance that the combination of the bent Laue and Bragg crystals allows an adaptation of the energy gradient without changing the focal length. Fig. 3 clearly shows the expected resolution minimum at the chosen reflection of an Fe calibration powder. 1 mrad of horizontal beam divergence was focused and the required energy gradient for the dispersion matching was 0.6%. The resolution was then limited by the beam size and the sample thickness. It could be improved by a larger sample-to-detector distance or by slits in front of and behind the sample confining the scattering volume. The ultimate resolution limit is set by the bandwidth of the Laue–Bragg monochromator.

2.3. *Vertical focusing*

Another useful feature of the meridional scattering geometry is the strict separation of the horizontal and vertical scattering planes. In a recent experiment on BM5, the Laue–Bragg monochromator was combined with a vertically focusing periodic laterally graded W/B_4C multilayer (Fig. 1). The elliptical bending was achieved by a two-momentum bender. The detailed parameters of the multilayer are given in Table 1.

The focus size was determined by a high-resolution film to be 4 μm, limited by the polishing errors of the substrate. It could be improved by a reduced focal distance which was deliberately chosen large to allow for a bulky sample environment (*e.g.* stress rig).

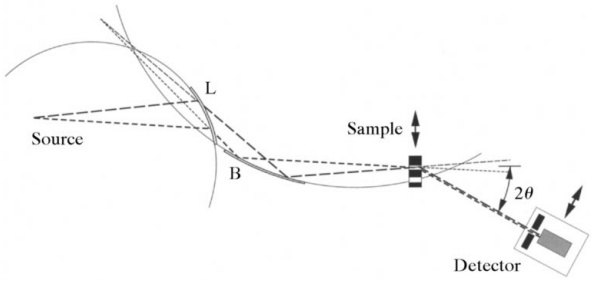

Figure 2
The Laue–Bragg monochromator in dispersion-compensating mode. For clarity of presentation, the multilayer which is present in Fig. 1 is omitted. An energy gradient across the focusing divergence is produced by increasing the bending radii such that the source lies off the Rowland circle. Beams indicated by long (short) dashes relate to low (high) energies. By dispersion matching, a parallel beam emerges from the powder-like sample.

3. Laue focusing

Slightly asymmetric bent Laue crystals in dispersive EXAFS geometry (Fig. 4) can provide microfocusing (Schulze & Lienert, 1996). The focal properties are best discussed in terms of the positions of the geometric and polychromatic foci. The geometric focus is formed by the beams emitted from a point source, each beam having the exact energy to be diffracted at the crystal surface. The focal distances are given by the lens equation. The polychromatic focus is formed as the energy width of each of the impinging beams is large compared with the bandwidth of the bent crystal. Each beam therefore spreads over the Borrmann fan and refocuses behind the crystal as described by the boundary conditions for reflection on bent asymmetric crystals. The formation of the polychromatic focus can be explained in a kinematical model describing the crystal bending by a change of the orientation and spacing of the reflecting lattice planes with the crystal depth. Therefore the diffraction angle of the beam penetrating into the crystal changes accordingly and a polychromatic focus is formed. A broadening of the focus is avoided if the geometric and polychromatic foci coincide, which can be achieved by choice of the asymmetry angle. The asymmetry angle depends on the energy. The

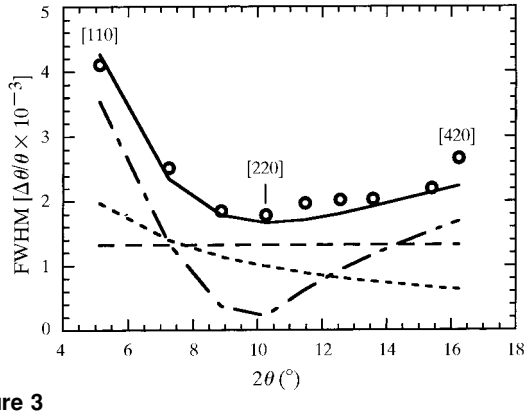

Figure 3
Resolution improvement by dispersion compensation. The measured peak widths (open circles) are plotted as obtained from a calibration Fe powder sample using dispersion compensation as shown in Fig. 2. The solid line indicates the resolution as calculated from the experimental parameters. Contributions to the total resolution arise from the sample thickness (long dashes), detectors slit opening (short dashes) and dispersion contribution (dashed-dotted). The minimum around the [220] reflection results from the dispersion compensation.

diffraction properties are discussed in more detail by Schulze *et al.* (1998).

The almost normal incidence of the Laue focusing geometry minimizes the beam path inside the crystal, resulting in a low absorbed power. The vertically diffracting Laue crystal was therefore operated in air without additional cooling. At ID15, 0.5×0.5 mm of direct beam were focused to a 1.2 μm-high horizontal line focus. The focal spot size was measured by fluorescence and absorption knife-edge scans. A 1 μm-thick lithographic Au test mask was used for the fluorescence knife-edge scans detecting the *K*-fluorescence with a Ge detector. The source size limitation of the focus size is 0.5 μm; the difference of the measured value could be due to various contributions such as alignment errors, inhomogenities of the knife-edge samples, vibrations or aberrations. The calculated efficiency of the bent Laue crystal is 90%. Laue focusing also provides low background and low tails arising only from the Compton and the thermal diffuse scattering in the thin crystal.

The Laue focusing geometry produces an energy gradient over the focused beam equal to $\Delta E/E = 1.1\%$ for 0.5 mm accepted beam. To compensate the prescribed dispersion, an analyser with tuneable dispersion, like a bent crystal, would be required.

4. Application: measurement of strain gradients in layered structures

Focused high-energy X-rays permit the use of transmission geometry to measure the residual strains in layered structures. The scattering geometry is shown in Fig. 5. Using focused low-energy X-rays, depth-resolved diffraction profiles might be obtained in Bragg geometry from cross sections but the residual strain properties will be biased by the existence of the artificial surface and by the preparation technique. Conventional and advanced techniques based on X-ray tubes, on the other hand, only provide cumulative depth information (see, for example, Genzel, 1996). The transmission geometry provides bulk and direct depth information. A narrow line focus fits the uniaxial sample geometry.

Table 2
Parameters of the microfocusing bent Laue crystal set-up.

The flux was measured using an Si PIN diode.

Energy	90 keV
Vertical source size (FWHM)	30 μm
Source–monochromator distance	65 m
Monochromator–focus distance	1 m
Reflection	[111]
Crystal thickness	1 mm
Asymmetry cut	0.42°
Bending radius	2 m
Intrinsic energy band width	5×10^{-4}
Aperture	0.5×0.5 mm
Geometric energy band width	1.1%
Focus size (H × V)	500×1.2 μm
Efficiency	90%
Flux	5×10^{9} photons s^{-1} $(0.1\ A)^{-1}$
Sample–detector distance	3 m

An accurate alignment of the sample tilts is required to avoid loss of the spatial resolution and artificial peak shifts at the interfaces. These peak shifts may arise as the effective beam centre of the scattered beam moves if a tilted line focus is scanned across an interface. The peak shift can be estimated as being half the length of the line focus, *i.e.* typically 200 μm. The absolute strain error is estimated as 10^{-3}. The measured samples had a width of 2–3 mm, requiring a tilt alignment better than 0.02° for a 1 μm focus size. The tilt angles were optimized by minimizing the width of absorption knife-edge scans on the sample surface. The focus size deduced from the sample knife-edge scans agreed closely with the results obtained by the fluorescence mask indicating a correct alignment. The above-discussed artificial peak shifts should therefore be confined to a 1–2 μm-wide region across the interfaces.

The potential of the technique is demonstrated for a Cu/Ni multilayer sample electrodeposited using the dual bath technique (Nielsen, 1996). The presented measurements were performed using the bent Laue focusing optics providing a spatial resolution of 1.2 μm. The sample

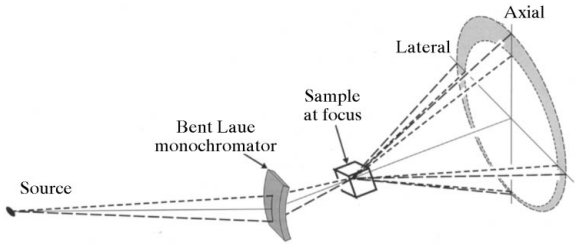

Figure 4
Sketch of the microdiffraction set-up on ID15. The vertical microfocusing was achieved by a bent Laue crystal. A Debye–Scherrer cone is shown. The varying width is due to the dispersion of the Laue monochromator. The beams indicated by long (short) dashes relate to low (high) energies. The parameters of the set-up are compiled in Table 2. Scintillation counters were employed and the scattering angle was measured by slit scans. The lateral and axial residual strain profiles were measured simultaneously using two detector/slit units.

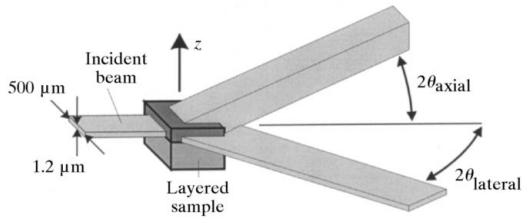

Figure 5
Transmission geometry for strain profiling on layered polycrystalline structures. The sample is scanned in the indicated *z* direction and the scattering angles are measured. The height of the line focus limits the spatial resolution to which strain gradients may be detected. Experiments were performed employing both the Laue–Bragg monochromator/multilayer combination and the Laue focusing optics to produce narrow line foci of 4 μm and 1.2 μm height, respectively.

structure and a contour plot of the [111] diffraction patterns are shown in Fig. 6. The broadening of the Ni peaks as compared with the Cu peaks is due to their smaller grain size. The [111] Ni layer reflections show an increased integrated intensity within a 10–15 μm-thick layer at the Cu interfaces, whereas the [200] reflections show a reduced intensity in the same region (*cf.* Fig. 7). This behaviour can be explained by a gradually changing texture. The peak centres were determined by least-squares fitting of pseudo-Voigt profiles. Fig. 7 shows the resulting relative change of the lattice plane distance, $\Delta d/d$, and the integrated peak intensities of the [200] reflection. The Ni layers show small $\Delta d/d$ gradients away from the interfaces and strong gradients within about 10–15 μm-wide regions next to the lower-lying Cu layers. These regions coincide with the regions where texture changes were observed. The signs of the $\Delta d/d$ gradients are reversed for the lateral and axial directions. The Cu signal is noisier, possibly due to the limited number of scattering grains, but it is clear that there is a difference between the average *d*-spacing of the Cu layers and the Cu substrate which also changes sign between the lateral and axial directions. Such inversion of signs is expected by the Poisson ratio coupling the lateral and axial strains. A change of the average lattice constant on the other hand would result in gradients of the same sign. It is therefore concluded that strain gradients must be present. The detailed evaluation of all measured reflections is in progress.

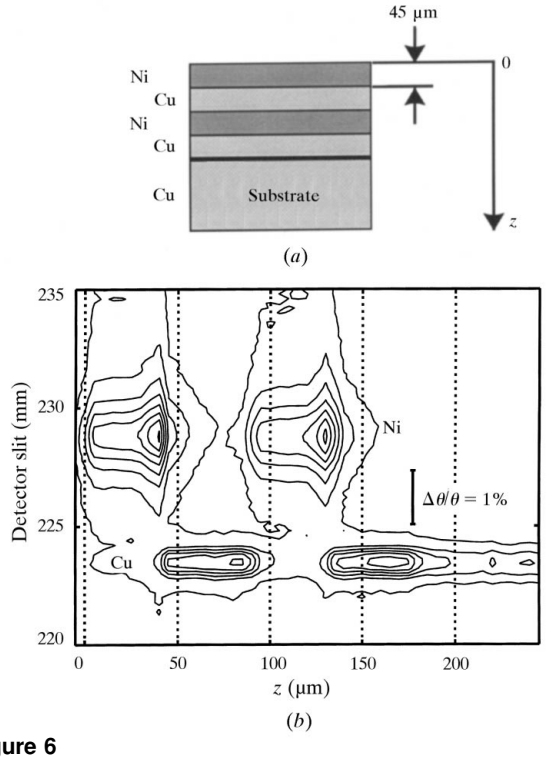

Figure 6
(*a*) Cross section of the Cu/Ni multilayer sample. (*b*) Contour plot of the axial [111] diffraction pattern from the sample using the bent Laue focusing optics (*cf.* Figs. 4 and 5). Plotted is the square root of the intensity *versus* depth and detector position. The contour spacing for the Cu layers is twice the spacing for the Ni layers. The detector position relates directly to the scattering angle as indicated by the vertical mark.

5. Conclusion and perspective

The commissioning on ESRF beamlines showed that a combination of meridional bent Laue and Bragg crystals provides a practical focusing monochromator scheme for the hard X-ray regime (40–140 keV). The ability to accept beams with a large opening divergence ideally suits the Laue–Bragg monochromator for wiggler and bending-magnet sources. It will also be beneficial for small-gap undulators on low-β sections, which are the best choice for high-energy focusing due to the small source size. It was demonstrated that broadening of the diffraction peaks due to the focusing divergence of the Laue–Bragg monochromator may be avoided by dispersion compensation. Furthermore, by combination of the Laue–Bragg monochromator with a curved multilayer, a 4 μm-wide line focus was obtained, exploiting the separation of the horizontal and the vertical scattering planes. This allows us to perform diffraction experiments with a high spatial resolution. If a supermirror is employed instead of the periodic multilayer, fixed-exit geometry is achieved also in the vertical direction. Then, spectroscopy becomes feasible where the narrow line focus allows the use of efficient high-resolution analyser crystals.

A 1.2 μm-high line focus was obtained using a bent Laue crystal. The Laue geometry provides almost normal incidence of the incoming beam, minimizing the aberration effects. Also, a very low background and focal tails are achieved. Applications can be hampered by the polychromatic geometry but it was demonstrated that the resulting energy bandwidth should be tolerable in many diffraction applications on polycrystalline samples and the

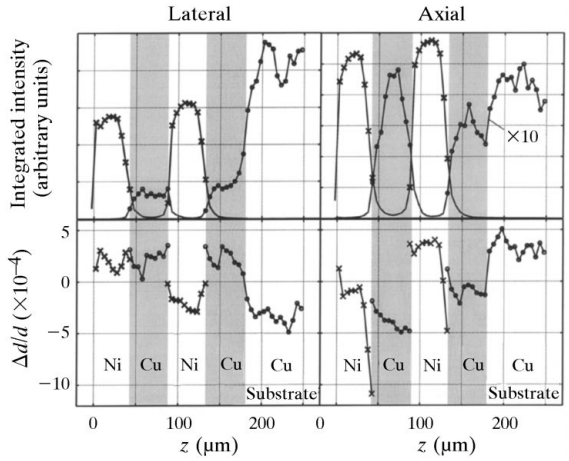

Figure 7
Relative *d*-spacings as evaluated from the [200] reflections along the axial and lateral directions (lower plots). The statistical error ($\pm\sigma$) from the peak fitting equals the symbol size at the centre of the layers and becomes about twice the symbol size at the interfaces. The integrated peak intensities are plotted above.

resolution can be improved by dispersion compensation. Two-dimensional focusing is achievable by combination with a curved multilayer. Such a combination can provide smaller focal spot sizes than the Laue–Bragg monochromator/multilayer combination, which in turn can provide monochromatic beam and more flux due to the larger aperture.

The microfocused hard X-ray beam was utilized to measure the strain gradients in layered polycrystalline structures in transmission geometry. For the first time, buried interfaces were investigated. The experimental results suggest that steep strain gradients and microstructural changes coexist within about 10–15 μm-wide regions close to the interfaces. The unique possibilities of this geometry are the direct observation of depth profiles and the access to buried interfaces. An obvious improvement of the technique would be the use of position-sensitive detectors instead of scanning slits. The errors due to the sample alignment can be reduced by two-dimensional focusing optics, larger sample-to-detector distances and crystal analysers. Absolute residual strains can be measured by a comparison with calibration powder samples, avoiding the severe limitations that pronounced texture poses on conventional methods (Genzel, 1997).

We anticipate that microfocused high-energy synchrotron radiation will be a powerful and unique tool for the materials scientist, filling a gap between the existing neutron scattering and electron microscopy techniques. The development of X-ray optics is of crucial importance for this new field and bent crystals and multilayers were found to be particularly suitable optical elements.

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