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Graded X-ray Optics for Synchrotron Radiation Applications

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Using X-ray diffractometry and spectral measurements, the structure and properties of graded X-ray optical elements have been examined. Experimental and theoretical data on X-ray supermirrors, which were prepared by the magnetron sputtering technique using precise thickness control, are reported. Measurements on graded aperiodic $Si_{1-x}Ge_x$ single crystals, which were grown by the Czochralski technique, are also presented. The lattice parameter of such a crystal changes almost linearly with increasing Ge concentration. The measurements indicate that $Si_{1-x}Ge_x$ crystals with concentrations up to 7 at.% Ge can be grown with a quality comparable to that of pure Si crystals.

Keywords: X-ray optics; crystal optics; X-ray monochromators.

1. Introduction

Graded X-ray optics should be outlined among modern research programs for the development of advanced X-ray and VUV optics for synchrotron radiation. Multilayer supermirrors, Göbel mirrors, laterally and in-depth graded crystals are among such optical elements. The application of these optics in synchrotron radiation beams is at an early stage. The potential of graded optics as first elements in beamlines for ultra-high-resolution monochromators and fine-focusing optics is very high.

Depth-graded optics are useful for enhancing the integral reflectance of a dispersive optical element. The principle of these optics is the same as for low-energy X-rays or VUV when multilayer optics are used, and for high-energy photons when crystals are applied. Due to different lattice parameters (multilayer periods) within the depth of the structure, such an element is able to reflect light in a wider energy range. This principle is shown in Fig. 1(*a*). In comparison with the periodical crystal (multilayer) the energy reflection curve for the depth-graded structure has a larger width and, therefore, a higher integral reflectance. This behaviour is demonstrated in Fig. 1(*b*) where results of model calculations of the spectral reflectance for periodic and graded W/Si multilayers made by Erko *et al.* (1995) are shown. The regular period of the multilayer and the number of layers are the same in both cases. Periodic multilayers are designed to achieve the best spectral resolution. Successful examples of X-ray supermirror tests were reported recently by Høghøj *et al.* (1994) and Erko *et al.* (1995). The tested W/Si supermirrors showed a measured reflectance of more than 30% for energies from 5 keV up to 65.5 keV.

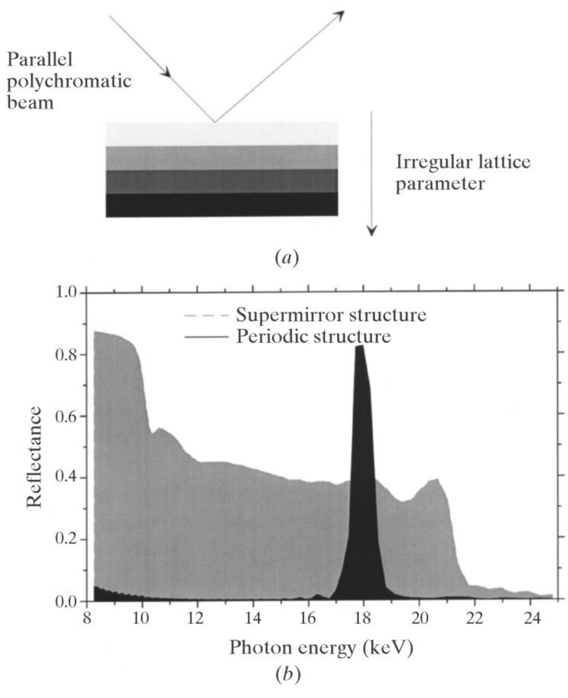

Figure 1
(*a*) Structure of a depth-graded optical element. (*b*) Spectral reflectance of a periodic (dark) and depth-graded (light) multilayer mirror in the case of a parallel polychromatic beam at a grazing angle of 0.32°.

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About 27 years ago Rustichelli (1970) suggested the application of depth-graded crystals in X-ray optics as dispersive elements with a spacing variation of the lattice parameter. Since then, there have been no successful examples of graded crystals because an extremely high distortion of the crystal lattice is needed to produce a sufficient gradient of the lattice parameter. Recently, several methods have been developed to produce crystals whose quality is comparable with those of perfect crystals. Depth-graded crystals have been successfully prepared and tested by Freund *et al.* (1972) as well as by Liss & Magerl (1994).

The feasibility of using optics with a lateral lattice gradient to modify the reflection properties has been reported by Nagel *et al.* (1982) for the case of multilayers and by Smither (1982) for the case of crystals. These optics can be used to enhance the spectral reflectance while the value of the integral reflectance remains constant. The principle of this type of optics is shown in Fig. 2(*a*). As an example, ray-tracing calculations of the spectral reflectance for a pure Si crystal and for an optimized graded crystal for the BESSY II beamline geometry with a naturally divergent beam, reported by Erko *et al.* (1996*a*), are shown in Fig. 2(*b*). Laterally graded multilayers can be easily fabricated with 200–300% period variations, so they can be used even with a conventional X-ray tube.

Interest in laterally graded crystals arose because of the development of new highly brilliant synchrotron sources. In particular, these elements can be of importance for third-generation storage rings with very small optical source sizes and vertical beam divergences of less than 0.2 mrad. The production of crystals with a lateral period variation is a much more difficult task. Due to internal defects a linear period variation of more than 0.04% cm^{-1} cannot be made. This fact limits the field of applications of laterally graded crystals to sources with very small divergences.

The physical principles of the crystal lattice parameter changes were investigated by Knapp & Smither (1986). A high-resolution monochromator system with a lateral gradient of the *d*-spacing by employing a thermal gradient has been discussed. Since then, several papers have been published describing the use of a crystal with a lattice spacing gradient as a focusing (imaging) element in X-ray and neutron optics (Bradaczek *et al.*, 1989, 1993). Successful attempts to grow KCl–RbCl laterally graded crystals were reported recently by Moshkin *et al.* (1997).

In this paper we present the properties of graded multilayers and SiGe single crystals. Graded multilayers were prepared by magnetron sputtering with a two-dimensional thickness control. The graded crystals were grown by the Czochralski technique on the basis of the $Si_{1-x}Ge_x$ alloy. The gradient was produced during the crystal growth by different concentrations of Ge in the Si matrix. The technique of growing such laterally graded crystals has now been successfully developed and first experimental results on their X-ray performance using soft

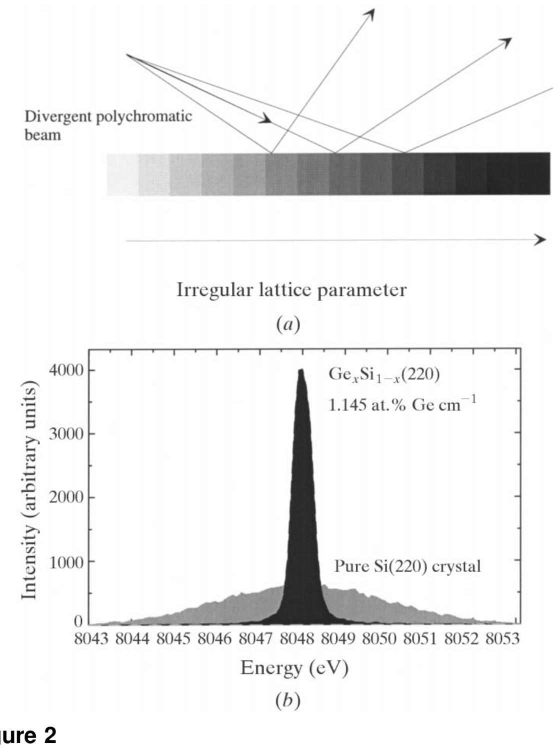

Figure 2
(*a*) Schematic structure of a laterally graded optical element. (*b*) Spectral reflectance of the pure (light) and laterally graded (dark) Si crystal in the case of a divergent polychromatic beam at a grazing angle of 23.651°.

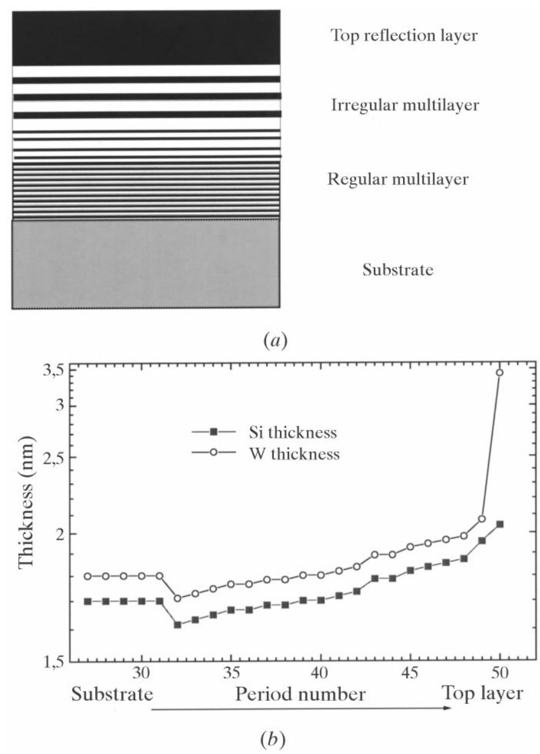

Figure 3
(*a*) Structure of a supermirror from the bottom to the top layer. (*b*) Period variation of the supermirror.

X-ray synchrotron radiation and X-ray diffractometry with Cu $K\alpha$ characteristic radiation are reported.

In addition, non-graded SiGe crystals with a rather low amount of Ge can be used to compensate the change of the lattice parameter which occurs during the cryogenic cooling of the crystals. Cryogenic cooling is, for example, necessary during the operation of the crystals as monochromators at synchrotron sources. First experiments with a cryogenic-cooled monochromator were performed at the ESRF by Souvorov *et al.* (1997).

2. Graded multilayer optics

To fulfil the conditions for the proposed BESSY II X-ray beamline, the bandpass of a supermirror must cover the energy range 5–20 keV at an angle of incidence of about 0.5°. Due to very high absorption at low energies, the number of layers constructively involved in interference is much smaller than for higher energies. This leads to considerable oscillations of the spectral reflectance in the defined spectral region. In order to smooth the spectral response, a combination of total external reflection from the top layer of a high-Z material with the interference reflection from a multilayer with a variable period has been applied. At first, from the substrate to the top, a regular multilayer with a period corresponding to the highest reflected energy is deposited. Then the multilayer structure with a variable period, and finally the thick layer of a heavy material, are deposited. The latter is chosen to provide total reflection conditions starting from the desired low-energy edge of the spectrum. The individual layer thicknesses were designed using software developed at the Laboratory of Electromagnetic Optics (LOE) at the University of Marseilles, France.

The supermirror has been fabricated in the same laboratory using the magnetron sputtering technique described by Vidal & Dhez (1988). W and Si layers are the optimum combination to fulfil the requirements for the designed energy range. The schematic structure of such a multilayer is shown in Fig. 3(*a*). In our experiment we used a period spacing in the range 3.5–7.0 nm on a 10 mm-thick silicon wafer as a substrate (Fig. 3*b*). The substrate roughness was measured as 0.2 nm r.m.s. The tungsten layer was deposited first; a total of 50 bilayers were prepared with variable spacing as described above.

The main goal of this design was to compensate for the decrease of the intensity of the synchrotron beam at higher energies in order to produce a beam with uniform spectral intensity from 5 keV up to 20 keV, as shown in Fig. 4(*a*). In Fig. 4(*b*) one can see a comparison between the calculated (dotted line) and measured (solid line) spectral reflectance for the designed supermirror. Measurements were performed by Neissendorfer from the University of Potsdam using the BESSY I wavelength shifter (WLS). The experimental data fit the theoretical predictions quite well.

3. X-ray optical elements on the basis of SiGe solid alloy

As is known, silicon and germanium form a continuous series of solid solutions with gradually varying properties. In Fig. 5 the variation of the lattice parameter going from pure Ge to pure Si is shown according to Dismukes *et al.* (1964). Evidently, a significant variation can be obtained with a very low concentration of Ge in Si. As can be clearly seen, the lattice parameter, d, changes almost linearly with increasing Ge concentration. A good approximation of the curve is given by Vegard's law,

$$\Delta d/d \simeq 4.18 \times 10^{-4}\, C, \qquad (1)$$

where C is the Ge concentration (in at.%).

The BESSY II storage ring is being designed for VUV and soft X-ray applications, but, by using insertion devices, such as a wavelength shifter or a multipole wiggler, beamlines up to approximately 50 keV photon energy are also available which will require hard X-ray optics.

Laterally graded crystals can be applied in high-resolution monochromators. For their application as monochromators, it is necessary to calculate the optimum lattice parameter variation. The result can be derived according

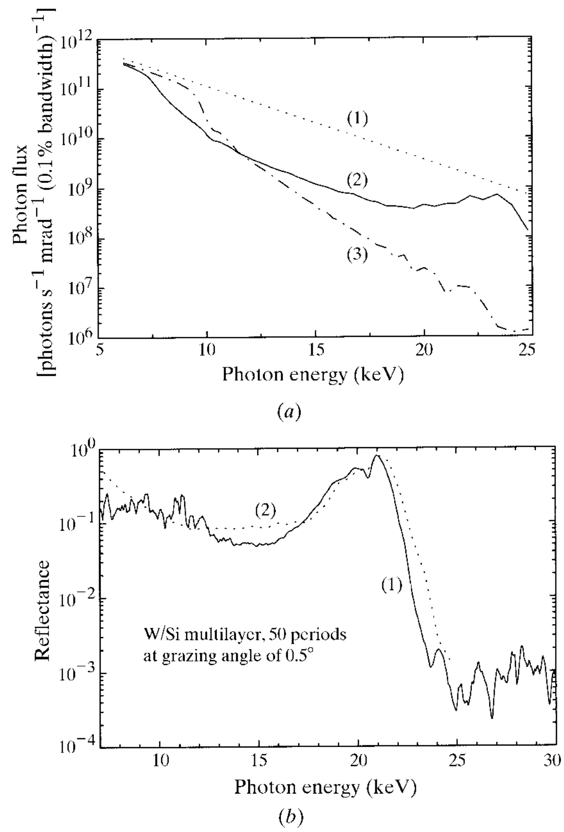

Figure 4
(*a*) Calculated spectral flux from the BESSY I wavelength shifter (WLS) beamline: (1) BESSY I WLS total flux, (2) supermirror at 0.5° grazing angle, (3) total reflection mirror at 0.5° grazing angle. (*b*) Spectral reflectance of the supermirror. Solid line: experimental data; dashed line: calculated data.

to Erko *et al.* (1996*a*) as follows,

$$G = 2.39 \times 10^3 \cos\theta_B / R, \quad (2)$$

where G is the concentration gradient (in units of at.% Ge cm^{-1}), R is the distance from the synchrotron source to the crystal, and θ_B is the Bragg angle. For all energies the resolution is given by

$$\Delta E/E = \left[\alpha_0(\cos\theta_{B_0} - \cos\theta_B)/\sin\theta_B\right] + (\Delta E/E)_n. \quad (3)$$

Using the parameter data of the BESSY II beamline one can effectively minimize the bandpass for one given energy E_0. In Fig. 6(*a*) the calculated energy resolution for two different gradient crystals optimized for two different energies is shown in comparison with a pure Si crystal. The maximum concentration gradient for the monochromator crystal, located at a distance of 20 m from the source, is equal to 1.23%. Using an asymmetrically cut crystal it is possible to decrease this value to 0.3 at.% cm^{-1}. It is clearly seen that for all energies away from the optimized energy the resolution is also considerably improved.

In addition to the improvement in energy resolution, a laterally graded crystal gives much higher spectral reflectance. The integral reflectance does not change, but the flux is concentrated in a smaller energy range. The gain in the spectral flux is approximately given by the ratio of the vertical divergence of the radiation accepted by the crystal and the Darwin width of the crystal,

$$g \simeq \psi/\Delta\theta_n. \quad (4)$$

As an example, the maximum achievable gain for Si(111) and Si(220) is plotted in Fig. 6(*b*), considering BESSY II dipole magnet radiation and an optimized Ge concentration gradient.

A dispersive or non-dispersive scheme can easily be realized in one design by simply inverting the direction of the gradient. Other possibilities can be thought of by introducing asymmetrically cut gradient crystals.

4. Experimental results

Single-crystalline $Si_{1-x}Ge_x$ ($0 < x < 0.1$) crystals have been grown by the Czochralski technique at the Institute of Crystal Growth (IKZ) in Berlin. Details of the growing process can be found in the paper by Abrosimov *et al.* (1996). A maximum Ge concentration of up to 7 at.% has been achieved along with a maximum concentration gradient of about 1.3 at.% cm^{-1} along the diffraction surface. [110]- and [111]-oriented specimens, which were polished on both sides, have been examined, with dimensions up to 70 × 50 mm.

Measurements of the lattice parameter variations were performed simultaneously at BESSY GmbH, at IKZ in Berlin, and at the Institute of Solid State Physics (ISSP) in Chernogolovka, Russia.

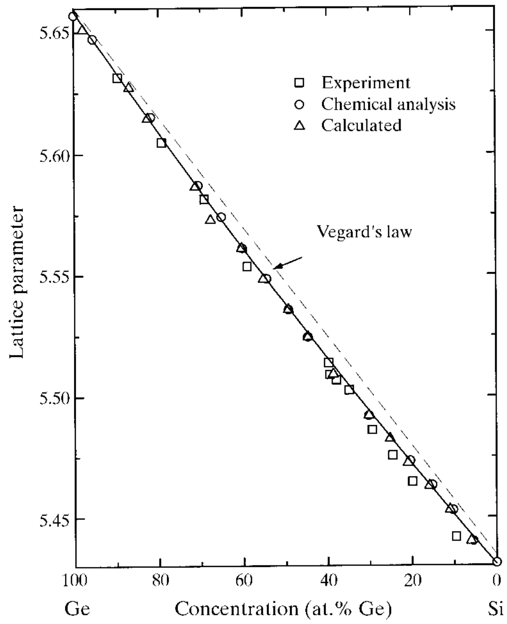

Figure 5
Dependence of the lattice parameter of the SiGe alloy as a function of the Ge concentration. The dashed line is an approximation according to Vegard's law.

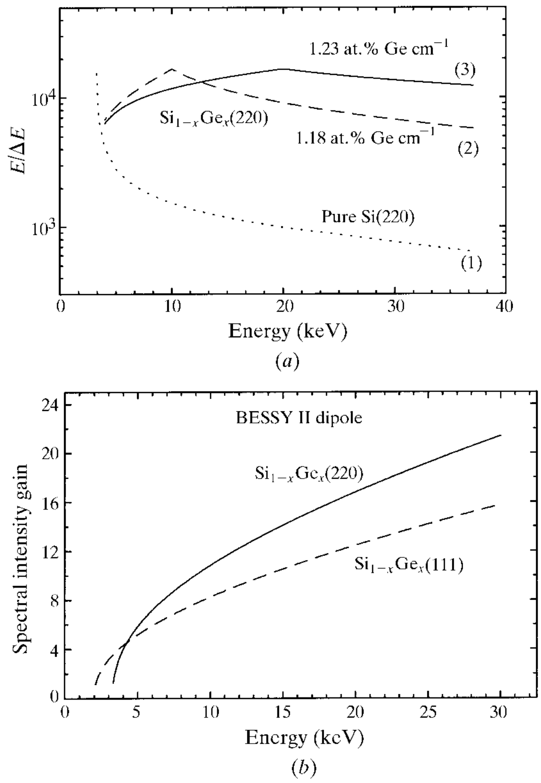

Figure 6
(*a*) Single-crystal energy resolution as a function of the photon energy for (1) pure Si crystal in a divergent beam, (2) gradient crystal optimized for 10 keV, and (3) gradient crystal optimized for 20 keV. All calculations are for BESSY II dipole radiation. (*b*) Maximum spectral intensity gain for the optimum gradient for two crystal reflections, (111) and (220), and for a pure Si crystal with corresponding orientations.

Lattice parameter measurements were performed using X-ray diffraction. This method is a very powerful tool for examining the variation of the lattice parameter of graded crystals since a shift of the large-angle Bragg maximum is involved with the change of the *d* value. Therefore, small variations of the lattice parameter can be derived with high precision. Exact measurements of the lattice parameter were performed at ISSP with an accuracy better than 0.1 at.% Ge in Si.

4.1. *Experimental set-up*

A scanning double-crystal X-ray diffractometer was used at BESSY for two-dimensional local lattice parameter measurements. A Philips-type X-ray tube with Cu $K\alpha$ radiation (λ = 0.154 nm) has been applied. The beam was monochromated with an asymmetrically cut Ge(220) crystal at an angle of incidence of 1.2°. Therefore, a highly collimated beam with an angular divergence of about 2 arcsec impinges on the sample. The horizontal beam size in the diffraction plane was about 100 µm. The sample holder can be translated automatically parallel and perpendicular to the incident beam in the range ±50 mm in the horizontal and ±10 mm in the vertical direction with a linear reproducibility of 10 µm. The angular step of the goniometer axis was 0.01 arcsec.

We are therefore capable of measuring the Bragg angle at different points of the sample and can derive the Ge concentration and the direction of the gradient by comparing the position of the maximum of the Bragg peak of the $Si_{1-x}Ge_x$ sample with the Si(111) or Si(220) reference position. Furthermore, the width of the peak permits some conclusions about the quality of the sample crystal in comparison with Si reference crystals. In addition, the absolute reflectance of the specimen can be measured, providing further information about the quality and the homogeneity of the crystal.

4.2. *Error calculations*

The random error is a very important feature of the goniometer, effectively reducing the preciseness and reproducibility of the obtained data. We have therefore performed the same scan 11 times on a well known Si(111) reference crystal. The angular position of the maximum was reproduced within a range of ±0.5 arcsec; in terms of Ge concentrations, this would mean that we can derive the Ge concentration of the sample with a preciseness of ±0.1 at.% Ge.

The systematic error due to surface curvature and scan angular deviation was calculated by the formulae described by Erko *et al.* (1996*b*). The difference in Ge concentration, ΔC, can be derived from two independent measurements of the Bragg peak positions for the different diffraction reflexes. For the [110] orientation one can write

$$\Delta C = 2.39 \times 10^3 \frac{\left[\Delta_{\rm exp}^{(440)} - \Delta_{\rm exp}^{(220)}\right]}{\tan\theta_{\rm B}^{(440)} - \tan\theta_{\rm B}^{(220)}}, \qquad (5)$$

(in units of at.% Ge) where $\Delta_{\rm exp}^{(440)}$ and $\Delta_{\rm exp}^{(220)}$ are the local shift of the Bragg angle for the (440) and (220) reflexes, respectively.

The surface curvature, $\Delta\alpha$, can be calculated using the formula

$$\Delta\alpha = \Delta_{\rm exp}^{(220)} - \frac{\left[\Delta_{\rm exp}^{(440)} - \Delta_{\rm exp}^{(220)}\right]\tan\theta_{\rm B}^{(220)}}{\tan\theta_{\rm B}^{(440)} - \tan\theta_{\rm B}^{(220)}}. \qquad (6)$$

From only one experiment we obtained information simultaneously about the Ge distribution and the sample curvature.

4.3. *Rocking-curve measurements and simulation*

Rocking curves for different sample orientations have been measured at an energy of 8 keV using an X-ray tube with Cu anode and an asymmetrically cut Ge(220) monochromator. All results obtained with graded samples were compared with reference silicon single-crystal diffraction curves under the same experimental conditions. For the rocking-curve measurements $Si_{1-x}Ge_x$ samples with uniform Ge surface concentrations of 1.5 at.% and 4.2 at.% were examined. The surface orientation of those crystals was [110]. The width of $Si_{1-x}Ge_x$ rocking curves for these crystals was the same as for a pure Si sample. In

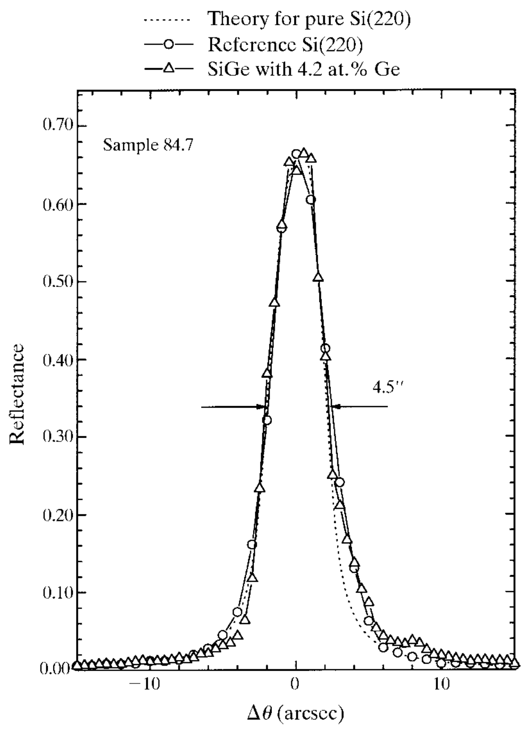

Figure 7
Rocking curves, obtained with Cu $K\alpha$ radiation, for a pure Si crystal and for the $Si_{1-x}Ge_x$ crystal with 4.2 at.% Ge in comparison with theoretical data.

Fig. 7 we present the rocking-curve measurements for an SiGe crystal with 4.2 at.% Ge in non-dispersive geometry and a pure Si crystal. Both crystals show a full width at half-maximum (FWHM) of 4.5 arcsec, which is only slightly above the theoretical predicted width of 4.2 arcsec for this configuration for a perfect crystal. The intensity of the beam reflected by the sample was normalized to the flux from the crystal monochromator.

4.4. *Lattice parameter variation measurements*

4.4.1. *Laterally graded crystal*. Samples for this experiment were cut along the growth axis; thus a concentration gradient was created along the surface. The variation of the lattice parameter was measured using a linear sample translation along and perpendicular to the X-ray beam of a small size. The beam size provided by the double-slit system was about 0.1×1 mm. The rocking curves of the (220) reflection of X-rays with 8 keV energy were measured in non-dispersive arrangement with a highly collimated beam at different positions on the sample along and perpendicular to the direction of the Ge gradient. Thus, the angular shift of the rocking curves corresponds to a local value in the lattice parameter, which was to be expected. The corresponding Ge concentration in the sample, as calculated from the angular shift of the rocking curves, is plotted in Fig. 8. The linear gradient of the Ge concentration along the surface in this case is about 1% cm^{-1}.

4.4.2. *Ge-doped Si crystals*. Samples for this experiment were cut perpendicular to the growth axis; thus a concentration gradient was directed perpendicular to the surface. In effect, a constant value of the Ge concentration along the surface has been obtained. The goal of these measurements was to test the uniformity of the Ge concentration perpendicular to the growth axis. For several samples we have found rather good uniformity in our experimental accuracy. One example is shown in Fig. 9, where we have plotted the Ge concentration distribution for the sample as a function of the coordinates on the surface. The average value of the Ge concentration is 4.2 at.% Ge.

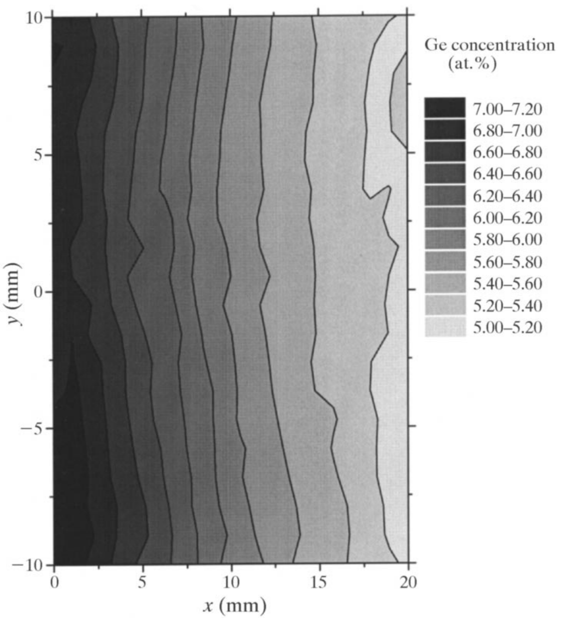

Figure 8
Concentration profile of Ge in Si calculated from the diffraction measurements for the SiGe graded crystal (Institute of Crystal Growth, Berlin, sample 39.27).

5. Conclusion

The application of graded multilayers and crystals as X-ray optical elements in synchrotron sources is very promising with respect to an increase of the energy resolution, an enhancement of the integral reflectance and the spectral flux depending of the gradient direction being used. Gradient crystals can be used in a divergent beam with almost the same energy resolution as that of a perfect Si crystal in a highly collimated beam. $Si_{1-x}Ge_x$ crystals without gradient could be used to compensate the thermal expansion coefficient of pure Si. A dispersive or non-dispersive scheme of monochromators can easily be realized in one design by simply inverting the direction of the gradient. Our first experimental data show that the quality and size of the $Si_{1-x}Ge_x$ crystals produced by the Institute of Crystal Growth, Berlin-Adlershof, is already sufficient for X-ray optics design.

We would like to express an appreciation to our colleagues from the Institute of Solid State Physics University of Potsdam for their cooperation in supermirror measurements. The growth of the gradient crystals was

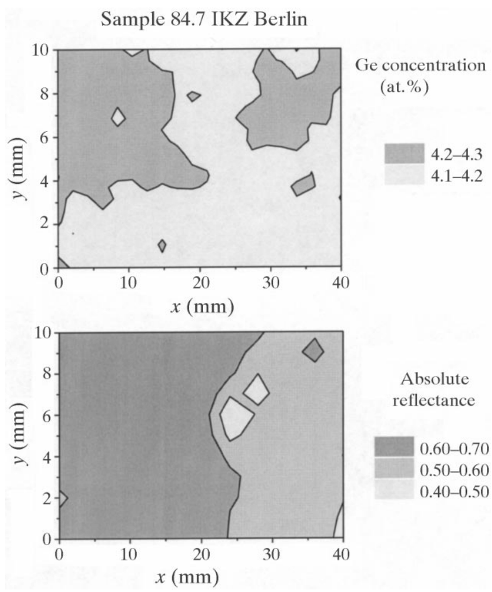

Figure 9
Concentration profile of Ge in Si calculated from the diffraction measurements for the doped crystal (Institute of Crystal Growth, Berlin, sample 84.7).

carried out in the Institute of Crystal Growth, Berlin, Germany, under support of the European Commission for the MONOCHESS II project (JOU2-CT92-0140). This work was supported by the Deutsche Forschungsgemeinschaft under contract AL 456/1-1.

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