$Si_{1-x}Ge_x$ Laterally Graded Crystals as Monochromators for X-Ray Absorption Spectroscopy Studies

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$Si_{1-x}Ge_x$ Laterally Graded Crystals as Monochromators for X-Ray Absorption Spectroscopy Studies

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Using the BESSY I wavelength shifter (WLS) beamline we have examined the energy resolution of a laterally graded $Si_{1-x}Ge_x$ crystal in the full divergent, white synchrotron beam. At the Fe-K absorption edge (E = 7112 eV) we have measured an energy resolution of $E/\Delta E=7.1\cdot 10^4$ for the (440) reflection and a vertical divergence of 0.63 mrad. For a pure Si(440) reference crystal and the same divergence we found $E/\Delta E=7.1\cdot 10^3$ which means a factor of 10 improvement in the case of the laterally graded crystal. Similar results have been obtained at the Co-K absorption edge (E = 7710 eV) with an increase of 2.6 for $E/\Delta E$. We present the experimental set-up and a method for the characterization of the absolute lattice parameter with a precision of $\Delta d/d = 2\cdot 10^{-5}$. In addition, we discuss the application of the laterally graded $Si_{1-x}Ge_x$ crystals in the crystal monochromator KMC-2 at the BESSY II storage ring.

KEYWORDS: x-ray diffraction, crystal optics, laterally graded crystals, $Si_{1-x}Ge_x$ crystals

1. Introduction

Crystals are widely used as monochromators for synchrotron radiation in the energy range from 1 keV up to 100 keV. In most cases, the energy resolution $E/\Delta E$ of a crystal monochromator is given by the divergence of the incident light. Therefore a collimating pre-mirror is required to obtain suitable energy resolutions. Such a mirror is very expensive and may reduce the available photon flux. Therefore, in 1986 Knapp *et. al.* [1)] suggested the use of a crystal with a variable lattice parameter along the surface to compensate the divergence of the incident beam. However, the realization of this type of crystal imposed significant problems because each distortion of the lattice reduces the quality and in effect the diffraction properties of the crystal. Only recently successful examples of the growth of laterally graded crystals have been reported. Those are crystals of the type $Si_{1-x}Ge_x$[2-7)], Bi-Sb[8)] and K-Rb biphtalate[9)]. Here, we examine $Si_{1-x}Ge_x$ single crystals grown by the Czochralski technique at the Institute of Crystal Growth Berlin[2,3)]. Differences in Ge concentrations along the Si crystal produce the lateral gradient of the crystal lattice parameter. The lattice parameter of the $Si_{1-x}Ge_x$ crystal as a function of the Ge concentration is shown in Fig. 1. According to Vegard's law[11)], the lattice parameter increases linearly with Ge concentration: $a_{SiGe} = a_{Ge}\cdot x+a_{Si}\cdot(1-x)$. As a rough estimation, this is a suitable approach. However, measurements on the lattice parameter revealed deviations from Vegard's law.

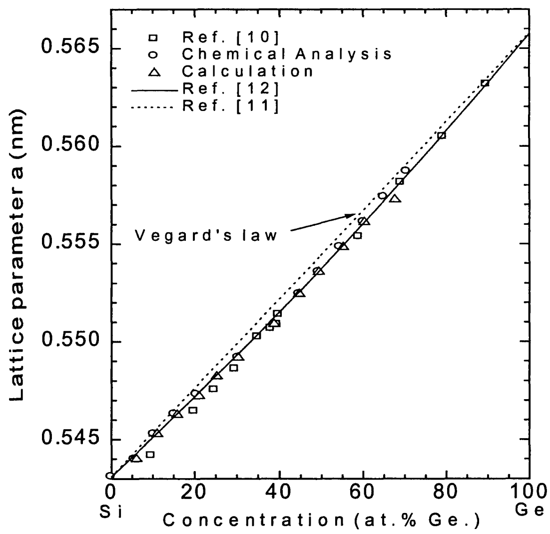

Fig. 1. The lattice parameter of $Si_{1-x}Ge_x$ crystals as a function of the Ge concentration according to Ref. 11. We would like to emphasize that there are no data points available in the concentration range $x \leq 0.1$.

A more precise description is given by the empirical formula $a_{SiGe} = (0.002733\cdot x^2 + 0.01992\cdot x + a_{Si})$ nm[12)]. For low Ge concentrations ($x \leq 0.1$) the distortion of the crystal lattice is rather small, therefore we expect the $Si_{1-x}Ge_x$

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laterally graded crystals to be perfect enough for applications.

2. Sample characterization with x-ray diffractometry

For measurements on the absolute lattice parameter variation, we used the Bond method[13] combined with two-dimensional scanning x-ray diffractometry[14]. For the error,

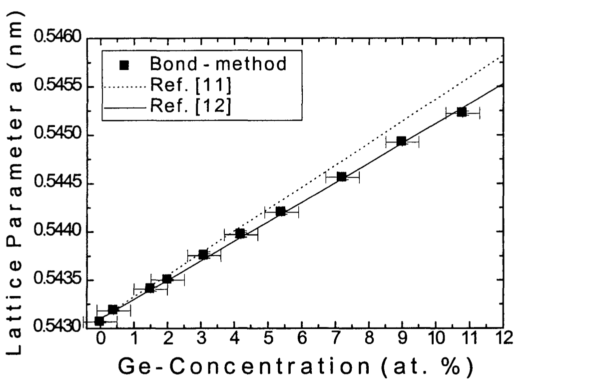

Fig. 2. Measurements using the Bond-method on the absolute lattice parameter of $Si_{1-x}Ge_x$ crystals. The Ge concentration was taken from infrared absorption spectroscopy measurements[15]. The deviation from Vegard's law is evident.

we found a value of $\Delta d/d=2\cdot10^{-5}$. With the Bond method it was possible to check the validity of Vegard's law experimentally for the concentration range $x \leq 0.1$ (Fig. 2). The empirical formula given in section 1 is well confirmed.

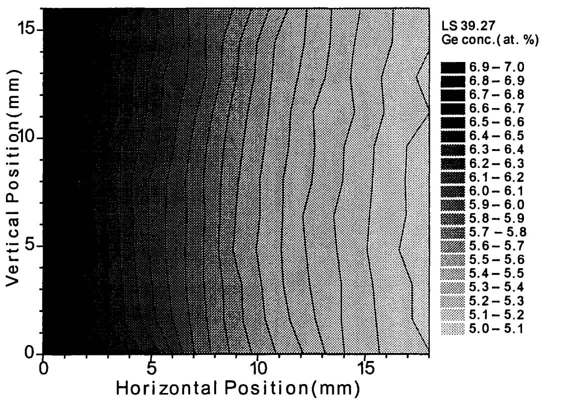

Fig. 3. Ge-concentration distribution of a $Si_{1-x}Ge_x$ (220) laterally graded crystal with a Ge concentration variation of 1.1 at. % per cm.

An example of a two-dimensional scan on a laterally graded $Si_{1-x}Ge_x$ crystal is shown in Fig. 3. As can be clearly seen, the lattice parameter variation is very smooth and almost constant, even close to the edges of the sample. For the concentration difference without surface curvature we obtain 2 at. % Ge, which perfectly matches values from infrared absorption measurements. From these measurements, the quality of the investigated gradient crystal is evident. The Ge concentration is relatively uniform in the direction perpendicular to the direction of the gradient. For the rocking curve width we have found similar results as for the reference Si crystal, indicating a low dislocation density and therefore an almost perfect crystal. The reflectance turned out to be homogenous and shows no systematic behavior as a function of the Ge content.

3. Experiment at the BESSY I WLS beamline

In advance of the use of the $Si_{1-x}Ge_x$ laterally graded crystals in the KMC - 2 monochromator it seems to be

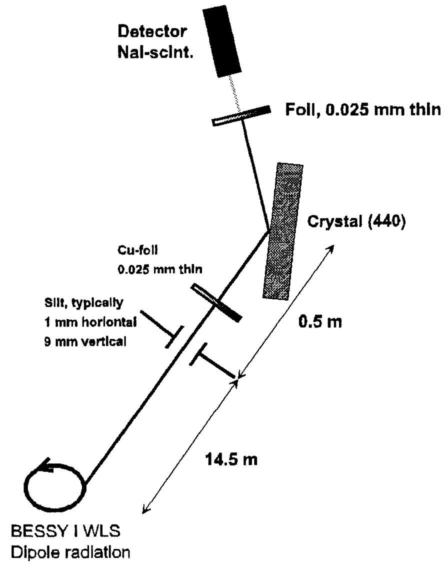

Fig. 4. Experimental set-up for the WLS Experiment. The vertical divergence is adjusted by the vertical entrance slit opening.

highly desirable to check their performance in a realistic situation. Therefore we built a single-crystal monochromator for the (440) reflection of $Si_{1-x}Ge_x$ and Si at the WLS beamline (Fig. 4). The white beam from the WLS passes through an entrance slit and is diffracted by the crystal. To determine the energy resolution of the crystal we used an energy scan through the absorption edges of Fe and Co. $E/\Delta E$ can not be measured directly with an energy dispersive detector since the energy width of the monochromatic beam is only about 0.1 eV at 7000 to 8000 eV.

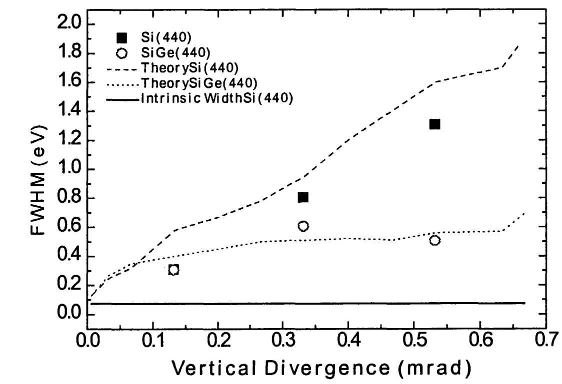

Fig. 5. Energy width of the reflected beam at the Co-K edge as a function of the vertical divergence, corresponding to the vertical slit width. Theoretical curves are ray tracing results.

In Fig. 5, we plot the energy width of the reflected, monochromatic beam as a function of the vertical divergence for the Co-K edge. The intrinsic width of Si(440), i.e. the energy resolution of a crystal in a parallel beam is 0.073 eV at this energy. Despite of the numerical disagreement between the ray tracing and experimental data due to the undetermined experimental error, the general tendency is evident. It can be clearly seen that the energy width of the pure Si(440) crystal increases almost linear with growing divergence as expected. In contrast, the energy width of the $Si_{1-x}Ge_x$ (440) laterally graded crystal remains nearly constant, independent of the slit width, i.e. the divergence of the beam. For 0.53 mrad divergence we found an energy resolution $E/\Delta E$ of $1.5 \cdot 10^4$ for the gradient crystal compared to $5.9 \cdot 10^3$ for the pure Si crystal at the same divergence of 0.53 mrad. Therefore, the energy resolution is improved by a factor of 2.6 as compared to 2.9 by theory. In conclusion, the theoretical predictions are reproduced quite well.

At the Fe-K edge with E = 7112 eV, we also made detailed measurements (Fig. 6). The intrinsic energy width is 0.068 eV in this case. At 0.067 mrad divergence, the graded crystal with the beam projection on the surface parallel to the gradient (down triangle) and non-graded crystal (up triangle) both show the same energy resolution. This is also predicted by ray tracing calculations although there is an offset in the absolute values. At 0.33 mrad divergence, we took one point with the graded crystal rotated by 180°, i.e. with the beam projection on the crystal surface anti-parallel to the direction of increasing Ge concentration. The effect is impressive.

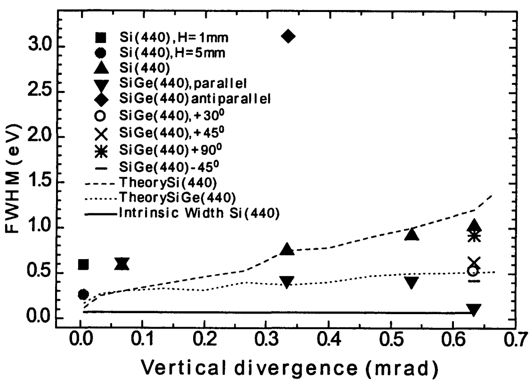

Fig. 6. Energy width of the reflected beam at the Fe-K edge as a function of the vertical divergence of the BESSY I WLS source. Theoretical curves are ray tracing results.

The energy width increases by a factor of 8 compared to the parallel case. The crystal acts now as a wide-band reflector, which clearly indicates that the gradient crystal in the correct position indeed compensates the divergence of the incident light. This improvement becomes clearer at 0.63 mrad vertical divergence, where we obtain a gain of the energy resolution of 10. If we rotate the gradient crystal by a certain angle α relative to the projection of the incident beam on the crystal surface, the gradient varies as $g' = g \cdot \cos\alpha$. In particular, if we rotate the crystal by 90° ($\cos 90° = 0$), there will be no gradient at all. The gradient crystal should then behave like a pure Si(440) crystal, which has in fact been observed within experimental uncertainties (the star in Fig. 6). We also tried other angles of 30° and 45°, which reduce the effective gradient and the energy resolution as expected.

3. The KMC-2 Monochromator at BESSY II

The potential of $Si_{1-x}Ge_x$ laterally graded crystals for the third generation synchrotron radiation source BESSY II have been described in the papers by Erko *et. al.* [16-18]. The results from the WLS experiment were very satisfying. Therefore we found it highly promising to use this type of crystal for a double-crystal monochromator called KMC-2 at BESSY II. The monochromator will be situated at a dipole source with the electron parameters horizontal source size 0.144 mm and vertical source size 0.041 mm. The beam divergence will be 0.370 mrad horizontally and 0.020 mrad vertically. Despite of apertures, slits and crystals no optical elements are present in the beamline.

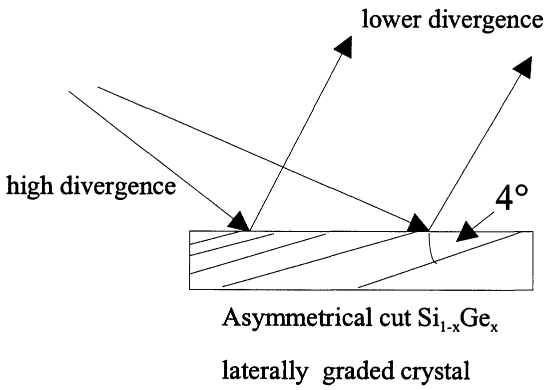

Fig. 7. The principle of asymmetrical cut gradient crystals. Due to the asymmetry, i.e. the constant angle of the crystal lattice planes towards the surface, the crystal acceptance is improved compared to the symmetrical case leading to a higher reflectance and lower divergence of the reflected beam.

We chose a special design for the monochromator. The first $Si_{1-x}Ge_x(111)$ crystal is asymmetrical cut with a constant angle of 4°. The advantages of this asymmetrical cut crystal are a higher reflectance and a collimation of the reflected, monochromatic beam (Fig. 7). The optimum gradient of the Ge concentration is 0.8 % Ge per cm for the

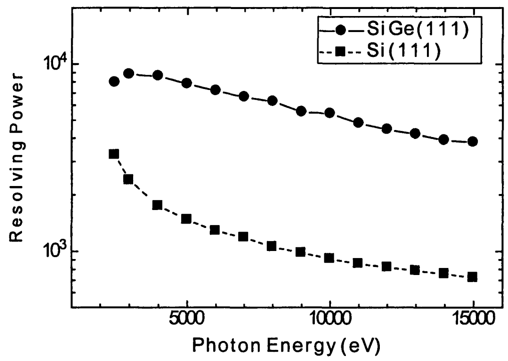

Fig. 8. Resolving power E/ΔE of the laterally graded crystal and the pure Si crystal in the divergent BESSY II beam.

first crystal at 10 keV photon energy. The energy resolution is determined by the second $Si_{1-x}Ge_x(111)$ crystal which is symmetrical cut and should have a gradient of 1.0 % Ge per cm for 10 keV photon energy in the ideal case.

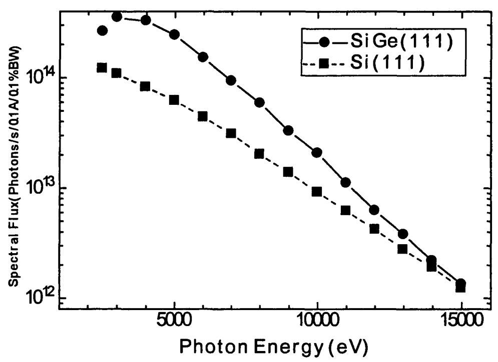

Fig. 9. Spectral Flux (i.e. Flux per Photon) for thepure Si(111) crystal and the laterally graded $Si_{1-x}Ge_x(111)$ crystal in the divergent BESSY II synchrotron radiation beam.

With this design, we expect an energy resolution and spectral flux as shown in Fig. 8 and 9. For comparison we also plot the performance of a double crystal Si(111) monochromator under the same experimental conditions. The energy resolution is also improved for energies far away from the optimum energy of 10 keV. The advantages of laterally graded crystals are evident. For further details we refer to the papers by Erko *et. al.* [16-18].

4. Conclusions

The advantages of laterally graded $Si_{1-x}Ge_x$ crystals are evident. They can be used in a divergent synchrotron beam with nearly the same energy resolution as a pure Si crystal in a highly collimated beam making them very attractive for high-resolution experiments.

The two-dimensional x-ray scanning together with the Bond method is a very fast and precise tool for the evaluation of the crystal properties. The measurements clearly revealed that the sample quality is comparable to commercially available Si crystals. The sample dimensions and value of the gradient are sufficient for applications at BESSY II. Our experiment at the WLS beamline confirmed the superiority of the laterally graded crystals. The energy resolution was improved by a factor of 2.6 at the Co-K absorption edge and by a factor of 10 at the Fe-K absorption edge. The experimental results are in good agreement with the theoretical predictions.

The first crystal monochromator beamline called KMC-2 using the $Si_{1-x}Ge_x$ laterally graded crystals is currently under construction at BESSY II and will be operational by the end of 1998. Measurements on the energy resolution and spectral flux in comparison to conventional crystals are on the way.

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