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Characteristics of PIXE channeling and its application to ZnSe thin films

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Abstract

The characteristics of the PIXE-method in channeling measurements of epitaxially grown ZnSe were studied and compared to RBS channeling. Especially the effect of the analysis depth and ion energy on the PIXE channeling minimum yield was focused on. The experimental minimum yields were compared to Monte Carlo simulations and the agreement was good. The PIXE minimum yield was found to increase significantly as a function of the ZnSe film thickness i.e. the analysis depth. A strong dependence of the channeling parameters on the beam angular divergence was also observed. According to our results, the PIXE minimum yield from ZnSe thin films does not follow the conventional energy dependence of the RBS minimum yield.

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1. Introduction

Rutherford backscattering spectrometry (RBS) combined with ion channeling is a widely used method for determining crystal perfection and lattice location of foreign atoms. However, RBS is not suitable for detecting neighbouring elements or light elements in a heavier matrix. In some cases a combination of RBS and nuclear reactions can be used. Suitable nuclear reactions are found, however, only for light elements. With particle induced X-ray emission (PIXE) it is possible to detect successive elements of the periodic table, for example zinc and gallium.

Lattice location studies require a comparison of yields from foreign and host atoms at the same depth. Unfortunately concentration depth information is difficult to obtain with PIXE. Because of this, minimum yields from foreign and host atoms are not readily comparable and a depth correction has to be made. Furthermore, when combinations of RBS and PIXE or nuclear reactions and PIXE are used, problems arise from the difference between close encounter probabilities in backscattering or nuclear reactions and the inner shell ionization probability for channeling directions.

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Mainly due to these difficulties only a few studies employing X-rays for lattice location determination have been published [1–8].

The purpose of this work is to investigate experimentally the characteristics of the PIXE-method in channeling measurements of epitaxially grown ZnSe, in particular, the dependence of the minimum yield on the analysis depth and ion energy. Some of these aspects have been studied theoretically by Comedi et al. [4], but to the knowledge of the authors no experimental studies of the subject have been done. We present new results of great importance especially when small differences in channeled yield have to be detected. This is the case, for example, in measuring asymmetric angular scans or in trying to detect interstitial host atoms.

2. Theoretical background

Two important parameters describe the crystalline structure of the sample or the lattice location of foreign atoms in ion channeling studies: the minimum yield $\chi_{\min}$ and the half width of the experimental angular scan $\Psi_{1/2}$. Minimum yield is the ratio of the channeled and random yields. For an axial direction, a semi-analytical expression for the RBS minimum yield originating just below the surface can be written as [9]:

$$\chi_{\min} = 2\pi cNdu_1^2\left[1+\left(\frac{\Psi_{1/2}d}{ku_1}\right)^2\right]^{1/2}, \qquad (1)$$

where

$$\Psi_{1/2} = C_\Psi\left[\frac{2Z_1Z_2e^2}{Ed}\right]^{1/2}. \qquad (2)$$

In these equations c is a parameter related to the angular divergence of the incident ion beam, N the atomic density, d the distance between atoms in a row, u_1 the one dimensional thermal vibration amplitude of the host atoms, and k a fitting parameter. Z_1 and Z_2 are the atomic numbers of the ion and the host atoms, respectively. C_Ψ is a parameter which depends on the vibration amplitude and E is ion energy. With values given in Ref. [9] for the parameters, Eq. (1) accurately describes the minimum yield in many cases. According to the equations, the width of the angular scan and the minimum yield decrease with increasing ion energy.

RBS and PIXE minimum yields are not readily comparable. Protons or helium ions moving along the crystal channels with energies higher than 100 keV typically approach the atomic rows to a minimum distance of about 0.1 Å. Since the impact parameter required for nuclear backscattering is of the order of 0.001 Å, there is a large reduction in the backscattering yield in channeling directions. Inner shell ionization is an atomic process and an ion can excite K-shell electrons with impact parameters higher than 0.01 Å. Because of this, PIXE minimum yield is always much higher and the width of the angular scan is smaller, than for RBS. In Monte Carlo simulations this can be taken into account by a method presented by Comedi et al. [4], where the basic idea is to replace the thermal vibration amplitude of the host atoms u_1 by $\sqrt{u_1^2+b_0^2}$, where b_0 is the mean impact parameter for the nner shell ionization.

Since the emitted X-rays in PIXE originate from the whole depth range of incident ions, dechanneling also strongly affects the obtained minimum yield. The deeper the ions travel in the crystal the larger is the dechanneling due to thermal vibrations of the crystal atoms and lattice defects. In Ref. [4] such depth correction factors to Eq. (1) have been approximated as well. In this paper, we have studied experimentally the effects of the analysis depth on the obtained minimum yield. The analysis depth was controlled by using epitaxially grown layers of varying thickness. Also the energy dependence of the minimum yield was studied.

3. Experimental

The ZnSe samples used in this study were grown at Tampere University of Technology by molecular beam epitaxy on an epitaxial GaAs buffer layer (on bulk GaAs). The undoped ZnSe layer thicknesses were 475–1890 nm. The measured RBS minimum yield was smaller than 6.0% for each of the samples indicating good crystalline quality.

The PIXE and RBS channeling experiments were carried out with 0.35–2.65 MeV $^{1}H^{+}$ ions obtained from the 2.5 MV Van de Graaff accelerator of the laboratory. The samples were mounted on a three axis goniometer having an angular resolution of 0.01°. Energy spectra of emitted characteristic X-rays were measured with an HPGe X-ray detector, which was placed at an angle of 135° with respect to the incident beam direction. The solid acceptance angle of the detector and the angular divergence of the incident ion beam were confined by slits and apertures to 1.1 msr and 0.015° or 0.030°, respectively. Backscattered particles were detected with a 50-mm^2 ion-implanted-silicon detector placed at a scattering angle of 170°. The ion doses were obtained by using an absolutely calibrated beam chopper system.

The thicknesses of the ZnSe layers and the crystalline quality of the samples were determined by RBS and channeling with 1.5–2.5 MeV $^{4}He^{+}$ ions. In these measurements the beam angular divergence was 0.015°.

4. Measurements and results

We used the Monte Carlo computer program Flux [10] to simulate the minimum yields. Flux is developed mainly for RBS channeling simulations, but has also been used in channeling studies involving nuclear reactions. In the PIXE simulations the Rutherford cross sections were substituted by K-shell ionization cross sections and the thermal vibration amplitude of the host atoms was corrected with the impact parameter b_0, as described earlier. For the ZnSe crystal no measured Debye temperature values are available, therefore the value was determined experimentally at room temperature from a 650 nm thick ZnSe sample. An RBS angular scan with 1.5 MeV ^{4}He ions from the $\langle 100 \rangle$ crystallographic axis was measured and compared to computer simulations with several different Debye temperature (T_D) values, assumed equal for zinc and selenium. The best fit was obtained with $T_D = 220 \pm 5$ K, corresponding to thermal vibration amplitudes of $u_1 = (0.117 \pm 0.003)$ Å and (0.106 ± 0.003) Å for zinc and selenium, respectively. In the following this value for the Debye temperature was used.

Channeling experiments are conventionally performed with protons and helium ions. The radiation damage induced by the analysing ^{4}He ion beam was studied by irradiating a ZnSe sample with 1.5 MeV ^{4}He ions, while the sample was aligned along the $\langle 100 \rangle$ axis. Almost a linear behaviour of the RBS minimum yield as a function of beam dose was obtained, as presented in Fig. 1. A clear increase in minimum yield would occur even for doses as low as 10 μC. From this we conclude that helium ions are not suitable when high doses are needed due to a small detector solid acceptance angle, as in the present setup.

Radiation damage can be reduced by using low mass ions, and therefore protons were adopted in this study. According to the theory by Kinchin and Pease [11] the cross section for atom displacement is proportional to the nuclear stopping power. The nuclear stopping power of ZnSe for ^{4}He ions is over 10 times higher than for ^{1}H ions in the present energy region. From this we can roughly estimate that 4 He ions produce about ten times more defects than ^{1}H ions. Accordingly, the resulting in-

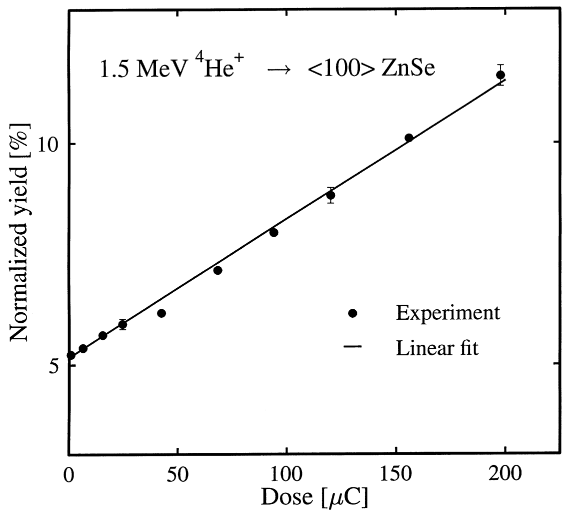

Fig. 1. The RBS minimum yield of ^{4}He ions incident on a ZnSe sample as a function of accumulated ion dose. The irradiations were done with 1.5 MeV ^{4}He ions with the sample aligned along the $\langle 100 \rangle$ crystallographic axis.

crease in the minimum yield would be negligible for the proton doses (10–15 μC) used in this study.

The difference between RBS and PIXE minimum yields was studied first; the results are shown in Fig. 2. The measurements were done from a 1750 nm thick ZnSe sample with proton energies of 800–2500 keV and with the beam angular divergence of 0.015°. All the PIXE yields in this study were taken from the zinc K_α peak. The analysis of the RBS spectra was done by using the GISA 3.99 computer program [12]. For stopping powers in the ⟨1 0 0⟩ channeling direction a value equal to 90% of the random direction value was adopted, in accordance with the preliminary results in Ref. [13]. Corresponding to the PIXE measurements the RBS yields were calculated from the whole ZnSe layer thickness. This way we eliminated the effect of depth dependence of the minimum yield, i.e., dechanneling. For the comparison of PIXE and RBS, these effects are not, however, completely eliminated because of the different energy dependences of the cross sections for the two methods. The general result can be seen in Fig. 2, where PIXE gives much higher minimum yields than RBS. It should be noted, that the RBS minimum yield in Fig. 2 does not correspond to Eq. (1), which involves the conventional definition of the minimum yield i.e. the minimum yield just below the surface.

The effect of a beam divergence smaller than 0.1° on the RBS minimum yield is negligible, as has been shown by Monte Carlo simulations [9]. However, in PIXE channeling even a small increase in the beam divergence strongly affects the minimum yield. This is depicted in the Fig. 3, showing the results of minimum yield measurements from a 1750 nm thick ZnSe sample with beam divergences of 0.015° and 0.030°. These measurements were done with energies of 650–2650 keV. Especially at energies higher than 1500 keV the difference is noticeable. According to our measurements the beam divergence is an important parameter in choosing the optimum ion beam energy.

To study how the PIXE minimum yield changes with the layer thickness, the minimum yields from ZnSe with thicknesses of 475, 650, 1250, 1750 and 1890 nm were measured. The measurements were done with proton energies of 500–2500 keV and with beam angular divergence of 0.030°. The results for energies 950 and 1100 keV are shown in Fig. 4. As may be noted, the agreement between experimental and simulated minimum yields is

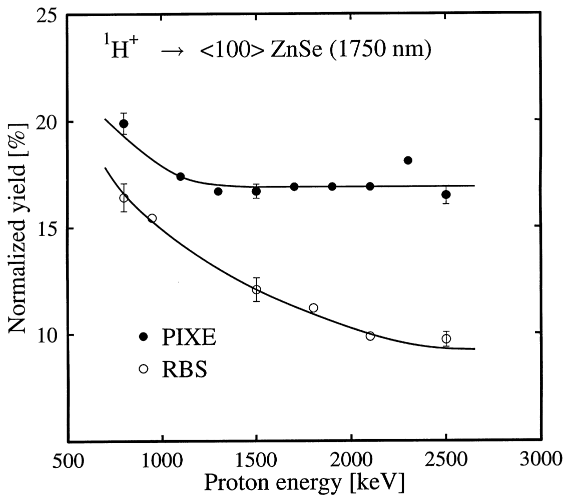

Fig. 2. The experimental RBS and PIXE minimum yields vs. incident proton energy from a 1750 nm thick ZnSe sample. The beam angular divergence was 0.015°. Solid lines were drawn to guide the eye.

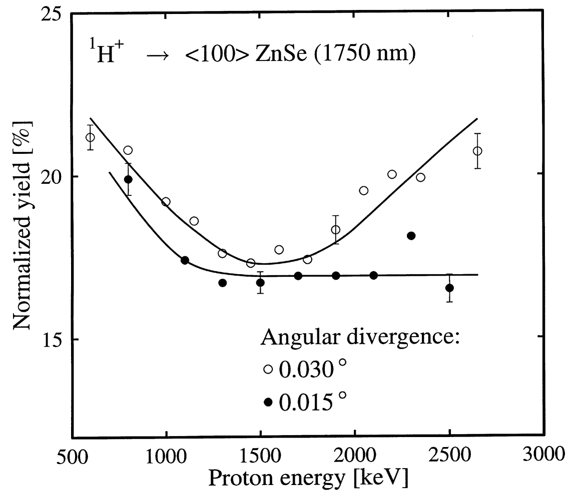

Fig. 3. The experimental PIXE minimum yields vs. incident proton energy from a 1750 nm thick ZnSe sample with beam angular divergences of 0.015° and 0.030°. Solid lines as in Fig. 2.

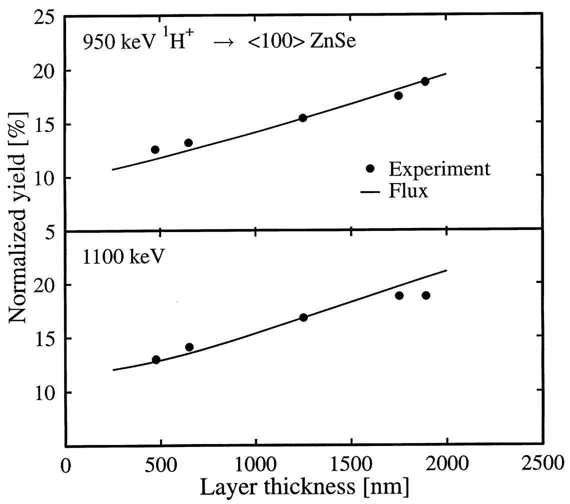

Fig. 4. The PIXE minimum yield vs. ZnSe layer thickness measured with proton energies of 950 and 1100 keV. The beam angular divergence was 0.030°. The experimental values are indicated by the points and the lines represent the values simulated with the code Flux. In this figure the statistical uncertainty of the experimental values is within the size of the symbols due to the scale of the *y*-axis.

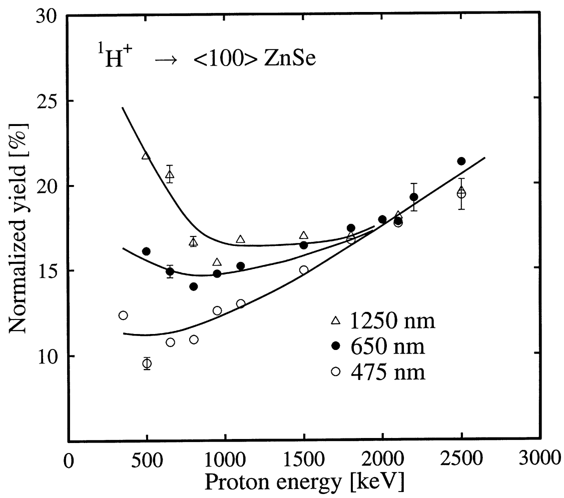

Fig. 5. The experimental PIXE minimum yield vs. incident proton energy from ZnSe samples with thicknesses of 475, 650 and 1250 nm. The beam angular divergence was 0.030°. Solid lines as in Fig. 2.

good. The minimum yield increases almost linearly as a function of the ZnSe layer thickness. The impact parameter values used in simulations for energies 950 and 1100 keV were $b_0 = 0.102$ and 0.117 Å, chosen by fitting to the experimental data. In general the maximum impact parameter is proportional to the square root of proton energy. The fitted values were about ten times the literature values of the impact parameter.

According to Eqs. (1) and (2) the subsurface RBS minimum yield decreases with increasing ion energy. This is also depicted in Fig. 2. The PIXE minimum yield, on the other hand, starts to increase above certain energy value, as shown in the Fig. 5. In this figure, the minimum yields for layer thicknesses of 475, 650 and 1250 nm are shown vs. incident proton energy. As may be seen, in the 350–1500 keV energy region, the minimum yield depends strongly on the layer thickness. At energies above 2000 keV there is no noticeable difference in the minimum yields for different layer thicknesses. A low minimum yield region may be observed in the curves in Fig. 5 and this seems to depend on the ZnSe layer thickness.

5. Discussion and conclusions

The angular divergence of the incident ion beam is an important parameter in PIXE channeling measurements. This effect has to be taken into account in analysis and in the choice of beam energy. The proper choice of beam energy is essential especially when small differences in the channeled yields have to be detected. As is the case, for example, in measuring asymmetric angular scans or in trying to detect interstitial host atoms.

Our experimental results show that direct comparisons between RBS and PIXE minimum yields are not feasible (Fig. 2). Even if the RBS yield is taken from the same depth interval, the PIXE channeling gives much higher minimum yield. If, for example, the angular scan from the host atoms is measured with RBS and the scan from the foreign atoms with PIXE, the special characteristics of PIXE such as presented in Figs. 2–5 have to be taken into account. From our experimental results it is possible to determine the magnitude of these effects in the case of ZnSe.

The analysis of PIXE measurements can best be done with computer simulations. In our Flux simulations the impact parameter b_0 was used as a fitting parameter including factors like the defects in the ZnSe layer which increase the minimum yield. Due to this the b_0 values do not correspond to the literature values. The effect of lattice defects on the minimum yield is similar to the effect of the increase of vibration amplitude.

As can be seen from Figs. 4 and 5 the PIXE minimum yield depends significantly on the analysis depth for proton energies of 350–1500 keV. Dechanneling depends inversely on the beam energy predicting smaller differences in minimum yields between different layer thicknesses at higher energies. This can be verified with computer simulations. A similar but stronger effect is shown by our results; there is no noticeable difference in the minimum yields at energies above 2000 keV. However, at higher energies the experimental minimum yield increases with increasing energy (Fig. 5).

One generally neglected phenomenon in PIXE channeling analysis is the secondary fluorescence. The significance of this phenomenon in the case of ZnSe/GaAs heterostructures was examined by comparing the energy dependencies of the minimum yields of zinc and selenium. A detectable difference was noticed. Thus, this effect might explain the above mentioned increase of the minimum yield of zinc at high energies. This effect will be studied in more detail in the future.

To conclude, the analysis of PIXE channeling is more complicated compared to the analysis of RBS and NRA channeling, but for many combinations of host and foreign atoms only PIXE can be used to get lattice site information.

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