Channeling of MeV ions in polyatomic epitaxial films: $ReSi_2$ on Si(100)

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Channeling of a He beam in the energy range from 1.4 to 2.7 MeV in a polyatomic epitaxial $ReSi_2$ film ($\sim$150 nm thick) was studied by detecting backscattered He ions. The critical angles and the minimum yields of both the heavy (Re) and the light (Si) elements are obtained directly from backscattering measurements. The critical angles of both Re and Si scale as $\sqrt{1/E}$. The critical angle of Re is always about 2.3 times that of Si. The minimum yields of both Re and Si do not change over this energy range. The minimum yield of Re (2%) is about $\frac{1}{7}$ that of Si (14%). The results are explained qualitatively and quantitatively by the continuum model suitably extended for polyatomic crystals. An important corollary is that a high value for the minimum yield of the light element in a polyatomic single crystal does not necessarily mean that the sublattice of the light elements is disordered.

I. INTRODUCTION

Channeling phenomena of MeV ions in crystals and their applications to materials characterization have been extensively reviewed.[1-3] Most experimental results on channeling can be understood in the framework of the continuum model established by Lindhard.[4] Two fundamental channeling parameters, the critical angle ψ_c and the minimum yield $\chi_{\min}$, have been thoroughly studied, for various ions (e.g., He and H) over a wide energy range (0.1–10 MeV) in numerous monoatomic crystals. For example, in diamond-type crystals, Picraux *et al.*,[5] found that the experimental half-angle (measured from angular scan) $\psi_{1/2}$ has the same functional dependence on parameters such as ion energy and atomic number as the critical angle ψ_c of the continuum model. Except for a few perfect crystals such as Si and Ge,[5] the measured minimum yields are usually larger than the predicted ones due to the existence of imperfections. The sensitivity of $\chi_{\min}$ to the defects in crystals makes it a good indicator of crystal perfection.

Channeling in polyatomic crystals has distinctive characteristics owing to the existence of distinct sublattices. The differences in channeling behavior from different sublattices are pronounced when the atoms occupying the sublattices have very different atomic numbers.[2,5] While these phenomena are of fundamental interest, for practical reasons, there have been relatively few experimental investigations of channeling in polyatomic crystals. For example, for bulk single crystals, it is difficult with backscattering analysis alone to detect the signal from light elements in the presence of heavy ones. One remedy is to measure the close encounter probability of an incident beam with the light element by detecting the characteristic x-ray production or nuclear reaction products from the light element in the channeling mode, while measuring the backscattered beam from the heavy element.[6-9] Another remedy is to use high-quality epitaxial thin films so that the signals from light and heavy elements do not overlap. We report here on a comprehensive and revealing experimental study of this type.

We have channeled a He beam (energy range 1.4–2.7 MeV) into an epitaxial $ReSi_2$ film grown on a Si(100) substrate. For a sufficiently thin film, the backscattering signal of the heavy (Re) element does not overlap in energy with that of the light (Si) element, which enables us to clearly separate the Si signal from the Re signal in the backscattering spectrum. Thus the half-angles and the minimum yields of both elements can be obtained from the backscattering measurements alone. This allows us to directly compare the channeling characteristics of different components of a polyatomic crystal. The experimental results are discussed in the framework of the continuum model, suitably extended for polyatomic crystals. The agreement is found to be good with the critical angles but only fair with the predicted minimum yields.

II. EXPERIMENTAL PROCEDURES AND RESULTS

$ReSi_2$ has a body-centered orthorhombic lattice with lattice constants of $a=0.311$ nm, $b=0.314$ nm, $c=0.767$ nm.[10,11] Its crystal structure closely resembles the well-known "$MoSi_2$" structure type. Since the difference between a and b is only $\sim$0.5%, we will hereafter assume for convenience that $ReSi_2$ is tetragonal with $a=0.313$ nm. The epitaxial $ReSi_2$/Si(100) structure was grown in ultrahigh vacuum by reactive deposition of Re on a Si(100) substrate held at $\sim$650 °C. Details of the

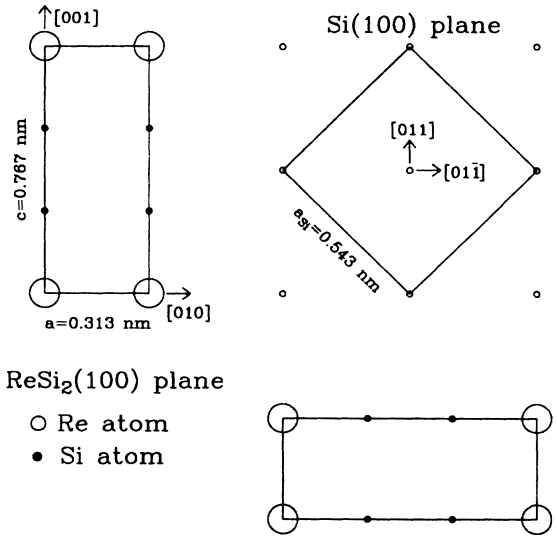

FIG. 1. The matching face of epitaxial $ReSi_2$ film and Si(100) substrate. Two distinct azimuthal orientations of the film with respect to the substrate are shown. The drawing is to scale. Only the orientations of the two $ReSi_2$ unit meshes with respect to the Si unit mesh are known; the actual positions shown are assumed.

growth procedures are described elsewhere.[12] The matching face of the $ReSi_2$ film and the Si substrate at the interface is $ReSi_2$(100)/Si(100) (Fig. 1). Two rotation twins with mutually perpendicular azimuthal alignments, $ReSi_2$[010]||Si[011] and $ReSi_2$[001]||Si[011] (Fig. 1),[12] exist because the Si(100) face has fourfold symmetry and the $ReSi_2$(100) face has only twofold symmetry. The sample for which results are reported here has a thickness of 150 nm and a composition of $Re_1Si_{2\pm0.1}$, as determined from 1.4-MeV He^+ backscattering spectroscopy (Fig. 2).

The channeling measurements were performed at room temperature with an x-y rotation and x-y translation goniometer. The vertical y rotation axis is fixed in space and perpendicular to the horizontal He beam. The x rotation axis lies in a horizontal plane and moves with the y rotation. The x-y translation directions are parallel to the corresponding rotation axes. First, the MeV He beam was aligned with the [100] axial channel of $ReSi_2$ by finding the minimum backscattering yield in a spectral window extending from beneath the surface peak of the Re signal to the energy corresponding to ~100 nm in depth. The same [100] axial channel was found by placing the window in the Si signal of the silicide or of the Si substrate instead of in the Re signal. This concurrence means that the [100] directions of the Si substrate and the $ReSi_2$ film are exactly aligned. Once the [100] channel had been identified, a channeling spectrum was then taken by translating the sample to a virgin spot (beam size $\sim 0.4\times0.4$ cm^2, sample size $\sim 2\times2$ cm^2) to minimize the effect of radiation damage.[13] Figure 2 shows the [100] axial channeling spectrum for a 1.4-MeV He^+ beam. One sees that in the $ReSi_2$ film, the minimum yield of Si much exceeds that of Re. The factor is seven—14% vs 2%. Channeling spectra were taken with several energies (1.4, 2.0, 2.4, 2.7 MeV) and the minimum yields are the same. This result is consistent with the continuum model which predicts that the minimum yield is independent of beam energy and is only a property of the host crystal.

The half-angles of the Re and the Si in the film and the Si in the substrate were obtained from angular scan measurements, which were performed by tilting the sample about either the x or y rotation axis and recording the backscattering yields within a spectral window corresponding to a thickness of ~100 nm placed in the proper region. The half-angles measured with a single channel just beneath the surface peak are the same as those measured with a spectral window, to within experimental error. To minimize the effect of radiation damage, we used a spectral window instead of a single channel for the an-

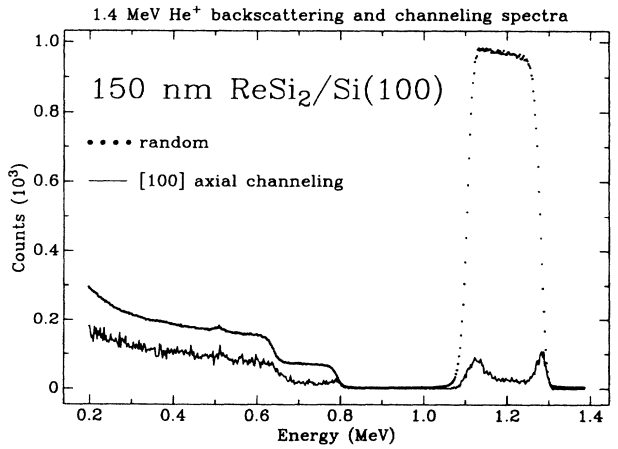

FIG. 2. 1.4-MeV He^+ backscattering spectra along a random direction (solid line) and a [100] aligned axial direction (dotted line).

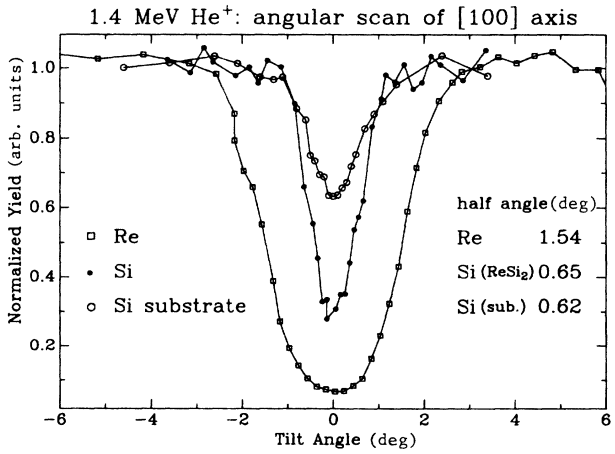

FIG. 3. A plot of normalized backscattering yield vs tilt angle. The normalization is performed with respect to the backscattering yield of a random incident beam. The half-angle is the half of the full width of the angular dip.

gular scan measurements, and we started each measurement on a virgin sample spot. The half-angles obtained by either x or y rotation are the same. Figure 3 shows the [100] angular scans for a 1.4-MeV He^+ beam. Two results immediately follow: (1) the half-angle of Re is ~ 2.3 times larger than that of Si in $ReSi_2$; (2) the half-angles of Si in the film and in the substrate are about the same. The half-angles were measured for several beam energies (1.4, 2.0, 2.7 MeV) with the same results, to within experimental error. Furthermore, all three half-angles decrease with increasing energy, proportionally to $1/\sqrt{E}$, as predicted by the continuum model.

III. THEORETICAL ANALYSIS

A. Theoretical background for channeling phenomena

The continuum model of axial channeling by Lindhard[4] represents the atomic chains along the channel of an elemental host crystal as continuous columns of radius $r_{\min}$ and electrostatic potential $U(r)$ where r is the distance from the axis of the column. $r_{\min}$ is the minimum distance a channeled ion may approach for the continuum model to still validly describe the gentle interaction between the ion and host crystal that keeps the ion channeled. There are two characteristic lengths: (1) the Thomas-Fermi screening distance, a_{TF}, and (2) the thermal vibrational amplitude of the host crystal ρ. Both are of the order of 0.01 nm in a typical channeling experiment. Lindhard has taken $r_{\min}^2$ to be the sum of a_{TF}^2 and ρ^2.

The meaning of $r_{\min}$ is that an ion of the incident beam aligned with a channel will be dechanneled if it impinges on the ends of the columns at the surface. Therefore, the minimum yield $\chi_{\min}$ measured beneath the surface peak for a perfect crystal is just the fraction of the surface area occupied by the columns

$$\chi_{\min}=\sigma\pi r_{\min}^2 , \tag{1}$$

where σ is the areal density of surface atoms.

The existence of $r_{\min}$ also implies that there is a critical angle ψ_c between an ion's trajectory and a channel above which the ion will be dechanneled,[4]

$$\psi_c=\alpha\left[\frac{r_{\min}}{a_{\mathrm{TF}}}\right]\psi_1 , \tag{2}$$

where the numerical coefficient α has a typical value between 0.6 and 1.6.[5] The characteristic angle ψ_1 equals

$$\psi_1\equiv(2Z_1Z_2e^2/Ed)^{1/2} , \tag{3}$$

where Z_1 and Z_2 are the atomic numbers of the ion and the host atom, E is the ion energy, and d is the atomic spacing in the channel direction.

B. Epitaxial $ReSi_2$ film as a vehicle for studying channeling in polyatomic crystals

In general, axial channeling in a diatomic crystal is complex because more than one type of column exist. However, there are special directions where channeling is simple. The simplest channeling direction has only one type of column which consists of both elements (e.g., the [001] channel in $ReSi_2$ for which we have no data). Channeling can then be viewed as being equivalent to that in a monoatomic crystal with an average atomic number and spacing.[5] The minimum yields and the critical angles of the two elements are then the same. The next simplest channeling direction has two types of column such as the [100] channel in $ReSi_2$ which we discuss in detail and for which we do present data.

The [100] channel consists of two types of column (Fig. 4); one type contains only Si atoms (Si columns) and another only Re atoms (Re columns). We assign a minimum distance of approach for each type of column, $r_{\min}^{\mathrm{Si}}$ and $r_{\min}^{\mathrm{Re}}$, which is determined by the characteristics of each corresponding column. This is the single column potential approximation.

For quantitative evaluations of minimum yields and critical angles, it is necessary to estimate the values of $r_{\min}$ in terms of channeling parameters of the ion and the host crystal. In general, $r_{\min}$ may depend on both the Thomas-Fermi screening distance[14]

$$a_{\mathrm{TF}}=0.047(\sqrt{Z_1}+\sqrt{Z_2})^{-2/3}\ \mathrm{nm} , \tag{4}$$

and the thermal vibrational amplitudes of the host crystal. For an incident He ($Z_1=2$) ion scattered by Si ($Z_2=14$) and Re ($Z_2=75$) atoms, the screening distances are 0.016 and 0.011 nm, respectively. For elemental Si and Re crystals, the known Debye temperatures $\Theta_D^{\mathrm{Si}}=550$ K and $\Theta_D^{\mathrm{Re}}=300$ K (Ref. 15) yield the thermal vibrational amplitudes of 0.011 and 0.008 nm, respectively, at room temperature, in the Debye approximation. These numbers are close to the values of the screening distances. To our knowledge, there is no reliable data on the thermal vibrational amplitudes in $ReSi_2$. Therefore, we shall approximate $r_{\min}$ by a_{TF} in computing the critical angles and the minimum yields.

In our single column potential approximation with $r_{\min}=a_{\mathrm{TF}}$, the critical angles can be directly calculated

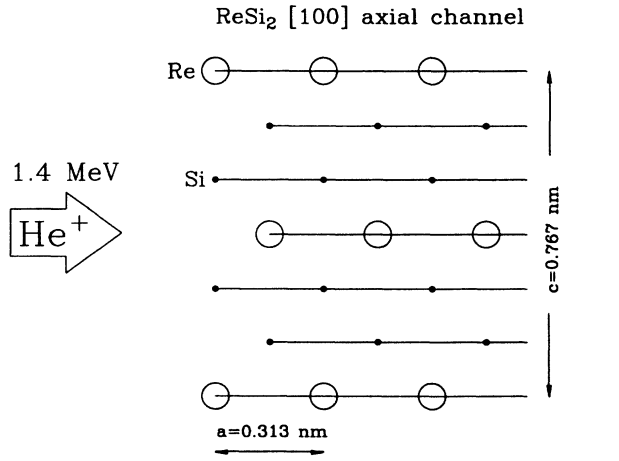

FIG. 4. A cross-sectional schematic diagram showing the [100] axial channel of $ReSi_2$.

TABLE I. Calculated values of the characteristic and critical angles, measured values of half-angles, and the calculated and measured minimum yields, for the [100] axial channeling of a 1.4-MeV He^+ beam in epitaxial $ReSi_2$ film.

	ψ_1 (deg)	ψ_c (deg)	$\psi_{1/2}^{expt}$ (deg)	$\chi_{min,1}$ (%)	$\chi_{min,2}$ (%)	χ_{min} (%)	χ_{min}^{expt} (%)
Si	0.78	0.65	0.65	3	10	13	14
Re	1.81	1.52	1.54	1	0	1	2

from Eq. (2). The numerical coefficient α equals 0.8 for both Re and Si columns using the "standard potential."[3] The ratio of the critical angle of the Re columns ψ_c^{Re} to the critical angle of the Si columns ψ_c^{Si} thus becomes

$$\psi_c^{Re}/\psi_c^{Si}=\psi_1^{Re}/\psi_1^{Si}=\sqrt{Z_{Re}/Z_{Si}}=2.3 , \quad (5)$$

which agrees with the experimental data (see Fig. 3). Table I lists the values of the characteristic angle ψ_1, the calculated critical angle ψ_c, and the measured half-angle $\psi_{1/2}$ for Si and Re in the $ReSi_2$ film. The measured half-angles agree well with the critical angles.

The minimum yields describes the fraction of an aligned incident beam that is dechanneled by atomic columns. An aligned beam impinging on $ReSi_2$ in the [100] direction will be scattered by both the Si and Re columns. The incident ion will be channeled by both Si and Re columns if the scattering angle ψ is less than the critical angles of both the Si columns and the Re columns. For the incident ion which is close to a Si column, the ion will be dechanneled by the Si columns but will still be channeled by the Re columns if the scattering angle ψ is greater than the critical angle of the Si columns ψ_c^{Si} but is still less than the critical angle of the Re columns ψ_c^{Re}. That dechanneling process thus does not contribute to the minimum yield of Re. Conversely, an incident ion which is close to a Re column will be channeled by Re columns but not by the Si columns if the scattering angle ψ is between the critical angles of the Si columns and the Re columns (i.e., $\psi_c^{Re}>\psi>\psi_c^{Si}$). That channeling process thus contributes to the minimum yield of Si. The minimum yield of the Si columns is therefore larger than that of the Re columns.

According to the previous reasoning, the minimum yield of Re columns χ_{min}^{Re} consists of two contributions, backscattering from Re atoms originating from the dechanneling when incident ions impinge on Re columns $\chi_{min,1}^{Re}$ and backscattering from Re atoms originating from the dechanneling when incident ions impinge on Si columns $\chi_{min,2}^{Re}$,

$$\chi_{min}^{Re}=\chi_{min,1}^{Re}+\chi_{min,2}^{Re} . \quad (6)$$

The values of these minimum yields can be estimated from appropriate modification of Eq. (1). Experimental results of channeling in elemental crystals indicate that Eq. (1) underestimates the value of the minimum yields.[3] Furthermore, Monte Carlo simulation of channeling phenomena by Barrett[16] indicates that the minimum yield extracted from computer simulation is about three times greater than that estimated from Eq. (1) and is in good agreement with the experimental data for perfect crystals such as Si and Ge.[3] We therefore modify Eq. (1) by a multiplying factor of 3 in order to get a better numerical estimation of the minimum yields. $\chi_{min,1}^{Re}$ thus can be estimated by

$$\chi_{min,1}^{Re}=3\sigma_{Re}\pi r_{min,Re}^2 , \quad (7)$$

where $r_{min,Re}$ is the minimum distance of approach to Re columns. To get $\chi_{min,2}^{Re}$, we need to know the approaching distance of incident ions to the Si columns r_{Si} when the scattering angle ψ equals the critical angle of Re columns,

$$\psi(r_{Si})=\psi_c^{Re} . \quad (8)$$

Knowing r_{Si}, we obtain

$$\chi_{min,2}^{Re}=3\sigma_{Si}\pi r_{Si}^2 . \quad (9)$$

The approaching distance r_{Si} obtained from Eq. (8) with the standard potential[3] is much smaller than $r_{min,Re}$ ($\sim 10^{-5}$ nm versus $\sim 10^{-2}$ nm) and hence $\chi_{min,2}^{Re}$ is negligible compared to $\chi_{min,1}^{Re}$ (see Table I). Therefore, one has

$$\chi_{min}^{Re}\approx\chi_{min,1}^{Re}\approx 3\sigma_{Re}\pi a_{TF,Re}^2\approx 1\% . \quad (10)$$

This predicted value of χ_{min}^{Re} is only about half the measured value. One explanation is that the $ReSi_2$ sample contains imperfections. It is not a single crystalline film. Another is that the approximation of r_{min} by a_{TF} is inaccurate.

Similarly, the minimum yield of Si columns χ_{min}^{Si} can be obtained,

$$\chi_{min}^{Si}=\chi_{min,1}^{Si}+\chi_{min,2}^{Si} =3\sigma_{Si}\pi r_{min,Si}^2+3\sigma_{Re}\pi r_{Re}^2 . \quad (11)$$

However, here the r_{Re} obtained from the equation

$$\psi(r_{Re})=\psi_c^{Si} \quad (12)$$

is larger than $r_{min,Si}$, so $\chi_{min,2}^{Si}$ is the major contribution to the minimum yield of Si columns. Table I lists the calculated and the measured values of the minimum yields for both Si and Re columns. Compared to the Re column, the relative good agreement between the measured value and the estimated value of the minimum yield for the Si column is probably due to its relative high value and hence its relative insensitivity to the imperfections in the film. This suggests that one should only use the minimum yield for the heavy (Re) element as an indicator of crystalline perfection, in particular in the case of low defects density.

IV. SUMMARY AND CONCLUSION

The continuum model, as extended here to polyatomic crystals, is able to explain the channeling phenomena that took place along directions for which there are more than

one type of columns as we have observed in $ReSi_2$. The critical angle of each column is determined by only the parameters of that column. The minimum yields are determined by the parameters of all columns, but are dominated by the dechanneling from the column with the largest average atomic number. These findings are readily generalized to channeling in polyatomic crystals with a number of different types of column, and from there to columns with different average atomic numbers and spacings. From the point of view of applying channeling to characterize the crystalline perfection of a polyatomic crystal, an important corrollary is that high crystalline quality is not synonymous with a low minimum yield for the light element. In particular, a high value of the minimum yield for the light element does not necessarily mean that the sublattice of the light element is disordered.

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