Radiation damage in $ReSi_2$ by a MeV ^4He beam

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(Received 17 May 1990; accepted for publication 16 August 1990)

Epitaxial $ReSi_2$ thin films grown on Si (100) substrates were analyzed at room temperature by MeV ^4He backscattering and channeling spectrometry. The minimum yield of [100] axial channeling increases with increasing exposure of the $ReSi_2$ sample to the analyzing He beam. This means that $ReSi_2$ suffers irradiation damage induced by a MeV ^4He beam. The damage in the film induced by a beam incident along a random direction is about one order of magnitude larger than that induced by a beam with an aligned incidence, indicating that the damage is mainly generated by elastic collisions of nuclei. The experimentally measured defect concentration produced at 300 K by a beam of random incidence is compared with the theoretically estimated one produced at 0 K in an amorphous target. The agreement is fairly good, suggesting that the defects are stable at room temperature.

Backscattering and channeling spectrometry are extensively used to probe compositional and structural properties of materials within submicron depths below the surface.[1,2] The effect of an analysis beam (of MeV ^4He or ^1He ions) on the materials studied is therefore important from both a practical point of view for correct interpretation of experimental data, and from a fundamental point of view for the understanding of MeV ion-solid interactions.

In the past two decades, there have been about a dozen studies explicitly concerned with the radiation damage of solids by MeV ^4He and ^1H beams.[3–13] Alkali halides are the most extensively studied class of materials,[3–6] where the ionization of target atoms by incident ions is the main mechanism for damage production. In Si and Ge, the damage is mainly produced by elastic nuclear collisions.[7–9] In GaP, both inelastic electronic ionization and elastic nuclear collisions contribute to the damage.[10] Extensive radiation damage has also been observed in some oxides [$BaTiO_3$,[11] NbO[12], Al_2O_3 (Ref. 13)]. Little work has been done on the radiation damage in transition-metal silicides by MeV ^4He and ^1H ions. Hensel *et al.*[14] studied the effect of 2 MeV ^4He irradiation on the resistivity of $CoSi_2$ and $NiSi_2$ thin films. Ishiwara *et al.*[15] used a MeV ^4He ion beam to analyze the radiation damage produced by 100 keV ^{40}Ar ions in expitaxial Pd_2Si and $NiSi_2$ thin films grown on Si substrates. Tsaur and Anderson[16] reported the increase of sheet resistance in silicides of Pt, Pd, and Ni upon 100 keV ^{40}Ar implantation.

We present here some experimental results on the damage induced by MeV ^4He ion irradiation in expitaxial $ReSi_2$ films thin grown on Si (100) substrates. Both the minimum yield of [100] axial channeling and the half angle were measured as a function of sample exposure to the ^4He analysis beam. The measured amount of damage produced by a random incident beam agrees with that computed from TRIM,[17] a Monte Carlo computer program which simulates the slowing down and scattering of energetic ions in amorphous targets. This agreement indicates that the total amount of damage produced by elastic nuclear collisions is preserved at room temperature.

An epitaxial $ReSi_2$ layer of ~150 nm thickness was grown on a hot Si (100) substrate (~650 °C) by "reactive deposition epitaxy" in ultrahigh vacuum ($\sim 10^{-19}$ Torr). Details of the growth procedure and characterization of the epitaxial $ReSi_2$/Si (100) structure are described elsewhere.[18] The fundamental parameters of channeling, the minimum yield, χ_{min}, and the critical angle, $\psi_{1/2}$ of the as-grown $ReSi_2$ (100) sample are discussed in a previous article.[19] Radiation damage produced by the analysis beam is the focus of this letter.

Experiments were performed at room temperature, using a MeV ^4He beam as both irradiation source and analysis probe, with the $ReSi_2$/Si (100) sample mounted on a goniometer with *x-y* translations and with two axes of rotation. To eliminate the effect of irradiation during the process of aligning the [100] channel with the incident beam, the channeling spectra were taken according to the following procedure: we first used the two rotation axes to find the [100] axial channel at one corner of the sample (beam size $\sim 0.2 \times 0.2$ cm^2, sample size $\sim 1 \times 1$ cm^2) and then translated the sample so that a virgin region of the sample was exposed to the irradiation beam for analysis. Figure 1 shows the backscattering spectra of the sample for a beam with random incidence (solid line) and for a beam incident along the [100] axial channel at three different damage stages: (a) as-grown (the dose during the channeling measurement of the as-grown sample is less than $\sim 10^{14}$/cm^2 and the damage induced is negligible), after irradiation by a 1.4 MeV $\sim 10^{17}$/cm^2 ^4He ion beam indicent; (b) along the [100] axial direction or; (c) along a random direction. Three facts are evident from the spectra: (1) the as-grown $ReSi_2$ sample is highly epitaxial with a Re minimum yield χ_{min} of ~2% (the fraction of counts below the surface peak of the aligned spectrum normalized with respect to that of a random spectrum); (2) substantial doses of the analysis beam produce damage in the $ReSi_2$

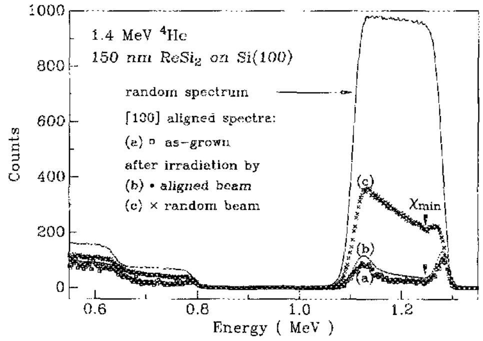

FIG. 1. 1.4 MeV ^{4}He backscattering and channeling spectra of a 150-nm-thick epitaxial ReSi$_2$ layer grown on a Si (100) substrate. All four spectra were taken at room temperature and are plotted by normalizing incident doses to a common value. The solid line is the spectrum for random incidence. The three [100] channeling spectra are for samples irradiated at room temperature with doses of (a) $\sim 10^{14}$/cm^2, $\sim 10^{17}$/cm^2, (b) in a [100] aligned direction, and (c) in a random direction.

film which results in noticeable increases of the minimum yield; and (3) the amount of damage produced by irradiation with an aligned beam is much smaller than that with a beam of random incidence. This last fact suggests that the damage is produced predominantly by elastic collisions among nuclei.

To further probe the damage structure of ReSi$_2$ by a MeV ^{4}He beam, we also measured the critical angles for the [100] axial channel before and after irradiation. In this channeling orientation, the atomic columns of the ReSi$_2$ lattice consist of only Si or only Re atoms. There are, therefore, two critical angles: one for Si columns and one for Re columns.[19] The angular scan and the critical angle of Re at the three damage stages discussed above (Fig. 1) are shown in Fig. 2. For the virgin sample, the critical angle for Re (as well as Si) agrees with Lindhard's prediction.[2] The critical angle decreases as the minimum yield (or damage) increases. It is known that disorder in the form of amorphous regions[20] or a mosaic structure[21] increases both the minimum yield and the critical angle. On the other hand, a spatially correlated disorder similar to that produced by lattice vibrations increases the minimum yield and decreases the critical angle.[8] The angular scan measurement on irradiated ReSi$_2$ (Fig. 2) therefore suggests a defect structure of correlated displacements for MeV ^{4}He irradiated ReSi$_2$. The same conclusion about the defect structure was obtained from the angular scans of the Si signal.

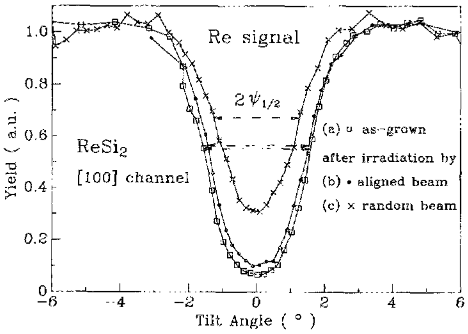

FIG. 2. Normalized backscattering yield of the Re signal vs angle of tilt between the incident beam and the [100] direction of the sample for the three damage stages of Fig. 1.

The damage induced by 1.4 MeV ^{4}He irradiation was quantified by monitoring the minimum yields of the [100] axial channeling spectra as a function of the total dose of exposure. To measure damage by a beam incident along a random direction, the sample was repetitively irradiated and then oriented in the [100] direction to take a channeling spectrum at increasing dose levels. Since a channeled beam generates little damage compared to that generated by the random beam (see Fig. 1), the channeling yield may be measured without significantly increasing the state of damage. The minimum yield χ_{min} of Si and Re initially increases rapidly (starting value: 14% and 2%, respectively) up to a dose of $\sim 2 \times 10^{15}$/cm^2 and then with a slower rate ($\sim 2.5\%/10^{16}$/cm^2 for Si and $\sim 1.9\%/10^{16}$/cm^2 for Re).

The fact that the minimum yield for Si is always larger than that for Re is peculiar to channeling of MeV ions in a diatomic crystal,[19] not an indication of higher initial or subsequent defect concentration for the Si sublattice. The reason is that the minimum yield of the element with low atomic number (Si) is enhanced and dominated by the ions deflected from the columns with the element of high atomic number (Re), while the minimum yield of Re is affected little by the deflection of He from Si columns.[19] We therefore use the minimum yield χ_{min} of Re as a measure of irradiation damage in ReSi$_2$.

To obtain the dose dependence of irradiation damage by aligned beam, we first oriented the sample in the [100] direction and then monitored the damage build-up by recording channeling spectra at increasing dose levels during irradiation. The minimum yield $\chi_{min}(\phi)$ of the sample after irradiation of dose, ϕ, is defined as

$$\chi_{min}(\phi) \equiv \frac{dN_A(\phi)}{d\phi} \Big/ \frac{dN_R(\phi)}{d\phi}, \tag{1}$$

where $N_A(\phi)$ and $N_R(\phi)$ are the total backscattering counts resulting from a dose ϕ of a [100] aligned and randomly incident beam respectively. $dN_A(\phi)/d\phi$ was obtained from channeling measurements by numerical differentiation. $dN_R(\phi)/d\phi$ is a dose-independent normalization constant, obtained from a random backscattering spectrum. Again, the minimum yield χ_{min} of Si and Re initially increases rapidly up to a dose $\sim 2 \times 10^{15}$/cm^2 and then with a slower rate ($\sim 0.9\%/10^{16}$/cm^2 for Si and $\sim 0.3\%/10^{16}$/cm^2 for Re). The rapid initial rise is difficult to grasp experimentally because of the poor statistics involved and will not be discussed further.

We used the TRIM88 program to simulate damage production by MeV ^{4}He ions in an amorphous 150-nm-thick ReSi$_2$ film on an amorphous Si substrate. The simulation computes the concentration (defect density/atomic density of ReSi$_2$) of displaced atoms as a function of depth in the linear cascade approximation.[17] A typical value for displacement threshold energy of 15 eV was chosen. The de-

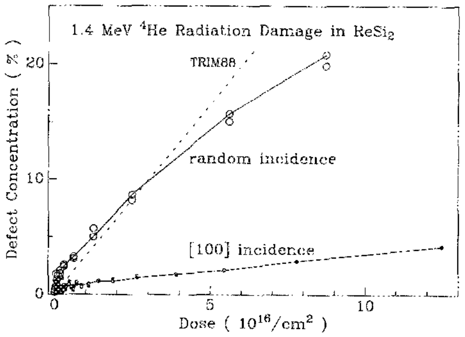

FIG. 3. Comparison of the measured defect concentration vs irradiation dose in ReSi$_2$ produced by 1.4 MeV ^{4}He ion beams of random (○) and [100] aligned (●) incidence at 300 K, and one calculated (dashed line) by a TRIM88 computer simulation of a beam of random incidence at 0 K.

fect concentration is roughly uniform in depth through the entire ReSi$_2$ film and is plotted versus dose in Fig. 3 (dashed line).

Experimentally, the defect concentration in a damaged crystal can be estimated from channeling measurements. When the defect concentration at depth z is $c_D(z)$ and the probability that an aligned incident beam is dechanneled by the defects over the region from the surface to the depth z is $P_D(z)$, we have[2]

$$c_D(z) + [1 - c_D(z)]P_D(z) = \frac{\chi_D(z) - \chi_V(z)}{1 - \chi_V(z)}, \qquad (2)$$

where χ_V and $\chi_D(z)$ are the normalized channeling yields at depth z for virgin and damaged crystals, respectively. In the near-surface region, the dechanneling probability P_D is small compared to the defect concentration c_D. Equation (2) therefore becomes

$$c_D = \frac{\chi_{\min,D} - \chi_{\min,V}}{1 - \chi_{\min,V}}, \qquad (3)$$

where $\chi_{\min,V}$ and $\chi_{\min,D}$ are the minimum yields. When the minimum yield of Re is used in Eq. (3), one obtains the defect concentration in ReSi$_2$ shown in Fig. 3. An aligned beam produced only about 1/7 the number of defects produced by a random beam. This is in accord with the observation that the close encounter probability between the incident ion and the target nuclei for an aligned beam is about one order of magnitude smaller than that for a random beam[2] and our assertion above that the defects are produced by elastic collisions among nuclei.

Figure 3 shows that the measured defect concentration produced by a random beam approximately equals that computed from TRIM88. We therefore conclude that the defects are stable at room temperature. This result is in contrast with that obtained for other silicides such as Pd$_2$Si (Ref. 15) and other semiconductors such as Si,[7] where the measured damage produced by light energetic ions (no dense cascade) at room temperature is much less than that predicted by TRIM88. The stability of defects may be explained by the semiconductor character and the relatively large cohesive energy of ReSi$_2$. Semiconductors are more sensitive to irradiation than metals because the strong chemical bonding in semiconductors gives a higher activation energy for vacancy-interstitial pairs to recombine. Defects are therefore more stable in semiconducting ReSi$_2$ than in metallic silicides such as Pd$_2$Si. In addition, ReSi$_2$ has a cohesive energy of 8.0 eV/atom (obtained from the cohesive energy[22] of elemental Re and Si and the heat of formation[23] of ReSi$_2$) compared with 4.6 eV/atom[22] for Si. This gives rise to a larger energy barrier for the migration of point defects in ReSi$_2$ than in Si. The sublinear rise of the measured defect concentration in Fig. 3 suggest that defects formed late in the irradiation are increasingly likely to be annihilated, resulting in a gradual saturation of the defect concentration as the damage increases.

This letter is based upon work supported in part by the Semiconductor Research Corporation under contract No. 100-SJ-89 and by the National Science Foundation under grant No. DMR-8811795 at Caltech, by the US Army Research Office and the National Science Foundation through grant No. ECS-8514842 at Colorado State University and by the National Aeronautics and Space Administration through a Small Business Innovation Research contract with the Colorado Research Development Corporation. The authors gratefully acknowledge this support.

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