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Swift Heavy Ion Beam-induced Recrystallisation of Buried Silicon Nitride Layer

T. Som^{1*}, O.P. Sinha², J. Ghatak¹, B. Satpati³, and D. Kanjilal⁴

¹Institute of Phsyics, Bhubaneswar-751 005 *E-mail: tsom@iopb.res.in ²AMITY Institute of Nanotechnology, Noida-201 303 ³ Institute for Minerals and Materials Technology, Bhubaneswar-751 013 ⁴Inter-University Accelerator Centre, New Delhi-110 067

ABSTRACT

Studies on MeV heavy ion beam-induced epitaxial crystallisation of a buried silicon nitride layer are reported. Transmission electron micrographs and selected area diffraction patterns have been used to study the recrystallisation of an ion beam-synthesised layer. Complete recrystallisation of the silicon nitride layer having good quality interfaces with the top- and the substrate-*Si* has been obsorved. Recrystallisation is achieved at significantly lower temperatures of 100 and 200°C for oxygen and silver ions, respectively. The fact that recrystallisation is achieved at the lowest temperature for the oxygen ions is discussed on the basis of energy loss processes.

Keywords: Ion beam-induced recrystallisation, silicon nitride, high-resolution transmission electron microscopy, selected area diffraction pattern, swift ion beam, SHI, swift heavy ion

1. INTRODUCTION

Silicon-on-insulator (SOI) structure is used in the integrated circuits of high packing density, low power consumption, and high speed¹. A device-worthy SOI structure should possess a silicon layer at the top, a good dielectric buried silicon nitride/oxide layer, and two high quality abrupt interfaces of the buried insulating layer with the top and the bottom *Si* layers for the fabrication of high-performance device. Crystalline silicon nitride is a wide band-gap material and the interface between crystalline silicon nitride and silicon is very stable.

SOI structures are typically synthesised by implantation of nitrogen/oxygen ions in silicon (at elevated temperatures), which gives rise to amorphisation, introduces roughness, and produces defects. On the other hand, device fabrication technology necessitates the removal of defects and recovery of the original crystalline structure. This is a rather complex problem because crystallisation of Si_3N_4 is an unsolved problem², and hence, a search is on to decipher a viable way to achieve recrystallisation of the buried layer with minimum defects.

Ion beam-induced epitaxial crystallisation (IBIEC) is a promising route to achieve solid phase epitaxial growth in silicon and other materials at considerably lower target temperatures³⁻⁹. IBIEC has advantages of low processing temperature and layer-by-layer crystallisation. For low to medium energy IBIEC, recrystallisation is attributed to the migration and recombination of defects (at the amorphous/ crystalline (a/c) interface) caused by the nuclear energy loss (S_n) process. On the other hand, recent works have demonstrated that swift heavy ions (SHI) can also induce recrystallisation in Si and Si_3N_4 , where the ions lose energy mostly by electronic energy loss (S_e) processes¹⁰⁻¹². Thus, SHI-induced ion beam-induced epitaxial crystallisation (SHIBIEC) may be considered as a subset of IBIEC.

In this paper, SHI-induced recrystallisation of an ion beam synthesised buried silicon nitride layer at temperatures as low as 100 °C has been reviewed. The results are explained in the light of the processes arising due to the electronic and the nuclear energy loss processes.

2. EXPERIMENTAL

Single crystalline Si(100) samples were first implanted with 100 keV N⁺ ions to the fluence of 8×10¹⁷ ions cm⁻² to form the buried nitride layer. Implantation was performed at 300 °C using high currents (30-40 µA cm⁻²). It can be mentioned that implantation performed using such high current densities causes the sample temperature to rise (self-annealing), which in turn leads to the recrystallisation of the top *Si* layer¹³. Monte Carlo SRIM-2006 simulation¹⁴ predicts the projected range of the nitrogen ions to be 245 nm.

The implanted samples were further irradiated by 100 MeV O^{8+} ions, 70 MeV Si^{5+} and 100 MeV Ag^{8+} ions at normal incidence to the constant fluence of 1×10^{14} ions cm⁻² at different temperatures, viz. room temperature (RT), 100, 150, 200, and 250 °C. The range of the projectile ions and the energy deposited by them were calculated by the SRIM-2006 code. The S_e and the S_n values of 100 MeV Ag ions in Si are given by 10.5 and 0.06 keV nm⁻¹, respectively, while those in the Si_3N_4 layer are given by 17.9 and 0.09

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keV nm⁻¹, respectively. Likewise, the S_e and the S_n values corresponding to 100 MeV O ions in Si are given by 0.7 and 0.0004 keV ^{nm-1}, respectively, while those in the Si_3N_4 layer are given by 1.1 and 0.0006 keV ^{nm-1}, respectively. Thus, the S_e/S_n ratio in the Si_3N_4 layer for both the ion species are given by 199 and 1833, respectively. Similar calculations for 70 MeV Si ions yield an S_e/S_n ratio in the Si_3N_4 layer to be 1115. Transmission electron microscopy (TEM) and selected area diffraction (SAD) patterns were used to examine the microstructure and the crystallinity of the samples. All cross-sectional TEM (XTEM) images were recorded at 200 keV using a JEOL-UHR2010 instrument.

3. RESULTS AND DISCUSSION

Figure 1(a) shows a low-magnification bright field TEM image obtained from the *N*-ion implanted sample. The presence of the implanted layer is clearly seen from the image show in Fig. 1. The crystalline nature of each layer was checked by collecting the respective SAD patterns, which are presented in Fig. 1(b). The SAD patterns indicate that the implanted layer is amorphous, while the top-*Si* layer and the substrate-*Si* are crystalline. The high-resolution TEM (HRTEM) images (not shown), corresponding to both the a/c interfaces indicate the presence of implantation-induced roughness.

TEM studies were also performed for the implanted samples irradiated further with Ag, Si, and O ions at RT. The SAD patterns (not shown) collected from all the three layers (as mentioned above) indicate that the implanted layer remains amorphous under SHI-irradiation at RT, while



Figure 1. Bright field XTEM micrographs obtained from an as-implanted (100 keV N^+ ions) sample: (a) low magnification image showing the three distinct regions, and (b) SAD patterns ([110] zone axis of *Si*) obtained from the three marked regions in (a).

the Si layers below and above it still remain single crystalline since it does not get amorphised under SHI irradiation with the present S_e values^{15,16}.

Fig. 2 shows a low-magnification TEM image obtained from an implanted sample, which was later irradiated by O ions at 100 °C. The SAD patterns obtained from the top *Si*- and the implanted-layer show that both are single crystalline in nature. It can be mentioned here that the SAD patterns were collected from the¹⁰⁰ zone axis of Si meaning that the HRTEM image of Si would show the d_{111} (=0.313 nm) spacing. The HRTEM image obtained from the marked region of the intermediate layer gives us a dspacing of 0.332 ± 0.004 nm, which is higher than the *Si*(111) plane spacing but close to the (200) plane spacing of the a- $Si_{3}N_{4}$ phase¹⁷. Thus, it can be inferred that a buried amorphous nitride layer is formed due to N-ion implantation, which gets fully recrystallised after 100 MeV O-ion irradiation. On the other hand, complete recrystallisation of the buried nitride layer is observed only at 200 °C for the samples subject to the silver-ion irradiation (using the same fluence). The author also observe complete recrystallisation of such a buried silicon nitride layer at 150 °C using 70 MeV Si ions¹⁰. It may be noted that the TEM results, corresponding to the Ag or the Si-ion irradiation, are not shown here because of the nearly similar features of the individual layers as described above (for the O ions).

From the above results, it is clear that recrystallisation of the buried silicon nitride layer takes place with O ions at 100 °C where the S_e/S_n ratio in the Si_3N_4 layer is 1833. Similarly, for the Si ions it occurs at 150°C where the S_i/S_n



Figure 2. Bright field XTEM micrographs obtained from an as-implanted sample, which was further irradiated with 100 MeV O^{8+} ions to the fluence of 1×10^{14} cm⁻² at 100 °C. The TEM image shows the top *Si*, the implanted layer, and the substrate *Si*. The dotted lines are a visual guide to demarcate the interfaces. The SAD patterns obtained from the three marked regions are also shown on the respective layers.

 S_n ratio in the Si_3N_4 layer is 1115. On the other hand, for Ag ions, although the S_e value in the Si_3N_4 layer 17.9 keV nm⁻¹, the S_n value is also reasonably high (0.09 keV nm⁻¹), which leads the recrystallisation of the nitride layer to take place at 200 °C (corresponding to the S_e/S_n ratio of 199). The above results indicate that the recrystallization of the buried nitride layer takes place for the O ions at a lower temperature, where the S_e/S_n ratio in the Si_3N_4 layer is much higher than that of Ag and Si ions. This raises the question of determining as to which factor dominates the recrystallisation process. Williams *et al.*⁴ have shown that the IBIEC in silicon occurs with low energy (0.6–3 MeV) N_e ions having much lower S_n (0.048–0.015 keV nm⁻¹) values compared to the Ag ions. Thus, the role of S_n cannot be completely neglected for the Ag-ion induced

recrystallisation of the Si_3N_4 layer. In fact, when the silver ions pass through the three layers and get buried into the substrate, they produce a lot of vacancies in the c-Si layers below and above the amorphous layer (~1.1 ion⁻¹ nm⁻¹ as obtained from SRIM-2003 simulations). Thus, at the fluence of 1×10^{14} ions cm⁻², a large number of point defects would be available near both the a/c interfaces.

In contrast, for the O (and Si) ions, the S_n values in the silicon and the nitride layer are very less leading to the generation of smaller number of vacancies in the topand the substrate-Si. Nonetheless, these vacancies migrate via thermal diffusion and the equilibrium sample temperature determines the diffusion coefficient. The vacancies will act like 'vacant spaces' in the amorphous layer and make the thermal vibrations of Si and N atoms (in the amorphous layer) relatively free and induce redistribution of the atoms, resulting in the crystallisation of one monolayer at the interface. Likewise, the recrystallissation process will progress from both sides of the amorphous Si_3N_4 layer.

The energy lost due through the inelastic scattering processes goes to the electron-hole pair production (through ionisation) and the excitation of electrons in the sample. The rapid energy transfer makes the system abnormally excited and the region around the ion track gets suddenly heated to a very high temperature within a small time scale¹⁸. A lot of excess vacancies (in addition to those formed by S_n and eventually survives) are created by this sudden temperature rise. Although these vacancies are produced all along the track, all of them cannot reach the interface and participate in the growth process. In fact, the typical number of vacancies to reach the a/c interface is $(\sim 10^{15} \text{ cm}^{-2})^{10,11}$, which help in enhancing the recrystallisation. Further, with the increasing number of the electron hole pairs, the neutral vacancies (activation energy for migration in *c*-Si (≈ 0.33 eV) are expected to get converted to doubly negative vacancies (activation energy for migration in c-Si ($\approx 0.18 \text{ eV}$) for dense electron hole pairs around them^{5,19}. Therefore, more the number of vacancies are produced by the incident ions; more doubly negative vacancies are formed. These move towards the a/c interfaces and lead to an enhancement in the recrystallization of the amorphous layer by continuous ion bombardment to a high fluence. Therefore, because of the S_{μ} -induced higher number of vacancy production, one would expect the Ag ions to promote the recrystallisation more efficiently than those of the O ions. However, a completely opposite trend is observed due to the reduced mobility of the large number of vacancies produced by the Ag ions, at lower temperatures. In contrast, the S_{μ} -induced vacancies produced by the O ions are almost negligible. Thus, they become easily mobile at relatively lower irradiation temperature of 100 °C to cause the atomic rearrangements.

Thus, in all the cases, although S_e is the dominant process and is much higher for the Ag ions, the highest S_e/S_n ratio for the O ions helps in accelerating the recrystallization of the buried Si_3N_4 at a much lower temperature. To find out a possible correlation, the respective recrystallisation



Figure 3. Variation in the recrystallisation temperature of the buried silicon nitride layer and the S_e/S_n ratio of different ions.

temperatures are plotted against the S_e/S_n ratios (in the silicon nitride layer) for all the three ion species (Fig. 3). These data show a linear relationship between the two and thus justifies our argument that higher the S_e/S_n ratio is lower is the recrystallisation temperature²⁰. Similar is the case for the SHIBIEC data available for *Si*, where it was shown that for different ions and energies, the higher S_e/S_n ratio leads to a higher regrowth rate for any given temperature²¹. It also shows the possibility of predicting the temperature at which the SHIBIEC sets in for the silicon nitride system.

4. CONCLUSIONS

In summary, the complete recrystallisation of a buried, amorphous silicon nitride layer due to Ag and O ions to the fluence of 1×10^{14} ions cm⁻². Oxygen ions lead to the recrystallisation at 100 °C, while the same is achieved at 200 °C for the silver ions. HRTEM studies indicate that the d-spacing of the recrystallised layer matches well with the a- Si_3N_4 phase. The observation that the oxygen ions lead to the recrystallisation at a relatively lower temperature is attributed to its higher S_e/S_n ratio in the nitride layer. The similar trend is observed for recrystallisation achieved for a similar sample using Si ions where the recrystallisation takes place at 150 °C corresponding to an intermediate S_e/S_n ratio. Based on these results, we propose that the S_e/S_n ratio is inversely proportional to the recrystallisation temperature.

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Contributors



Dr T. Som is an Assistant Professor at Institute of Physics, Bhubaneswar, India. His research interests include growth, characterization, and ion beam induced modification of metal, semiconductor, and insulators materials. These studies include ion beam induced epitaxial recrystallization, ion beam induced thin film/nanoscale magnetism, ion beam

synthesis and modification of nanostructures, and ion beam mixing of thin multilayers. He has published more than seventy papers in peer-reviewed international journals and edited volumes/books.



Dr O.P. Sinha is a Senior Lecturer at AMITY Institute of Nanotechnology, Noida, U.P, India. His research interests include swift heavy ion induced modification of semiconductors and alloys. He is also engaged in studying low-to-intermediate energy ion beam induced surface nanostructuring of alkali halides and semiconductors. He has special experience

to work with UHV atomic force microscope to characterize the nanoscale surface features of ion beam modified semiconductor surfaces. He has published twenty papers in peer-reviewed international journals.



Dr J. Ghatak is presently working as a Post-doctoral fellow at Dept of Material Science and Engineering at the National Cheng Kung University, Taiwan. His present research interests are to study the interfaces of II-VI semiconductor interfaces, the analysis of micro structure of II-VI semiconductor nanorods, and ion beam induced modification of materials.

He is well experienced in transmission electron microscopy. He has published nearly fifty papers in peer-reviewed international journals.



Dr B. Satpati is presently a scientist at Center for Advanced Material Processing, Central Mechanical Engineering Research Institute, Durgapur, West Bengal, India. His research interests include growth, modification, and characterization of nanodimensional metal, semiconductor, and insulating thin films. He has used energetic ion beams to modify thin film

nanostructures. He has a long experience of working with transmission electron microscope at different renowned institutes. He has published more than seventy research articles in peer-reviewed international journals.



Dr D. Kanjilal is a senior scientist and program leader at the Inter-University Accelerator Centre, New Delhi, India. He has a long research experience with laser and energetic ion beams. His present research interests include development of ion sources, accelerators, and materials modification by energetic ion beams. He has established a few unique facilities

at IUAC, which are being used by hundreds of university users. He has published more than 300 research articles in peer-reviewed international journals and also edited a few books and conference proceedings.