Monte Carlo simulation study of a selforganisation process leading to ordered precipitate structures

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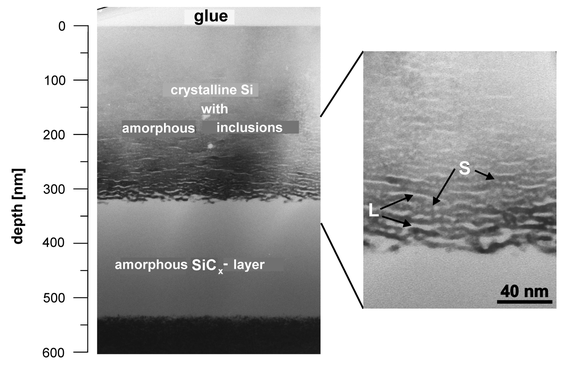
Motivation

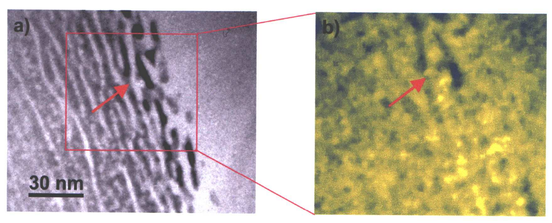
Experimentally observed selforganisation process at high-dose carbon implantations under certain implantation conditions.

- Regularly spaced, nanometric spherical and lamellar amorphous inclusions at the upper a/c interface



Cross-section TEM bright-field images:
$180\,keV\ C^+ \rightarrow Si$, $T_i = 150\,°\mathrm{C}$, Dose: $4.3 \times 10^{17}\,cm^{-2}$
Amorphous inclusions appear white on darker backgrounds
L: amorphous lamellae, S: spherical amorphous inclusions

- Carbon accumulation in amorphous volumes



Bright-field TEM image and respective EFTEM C map:
$180\,keV\ C^+ \rightarrow Si$, $T_i = 200\,°\mathrm{C}$, Dose: $4.3 \times 10^{17}\,cm^{-2}$
yellow/blue: high/low concentrations of carbon

Similarly ordered precipitate nanostructures also observed for a number of ion/target combinations for which the material undergoes drastic density change upon amorphisation.

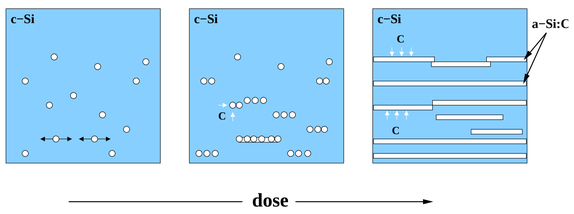
A. H. van Ommen, Nucl. Instr. and Meth. B 39 (1989) 194.

E. D. Specht et al., Nucl. Instr. and Meth. B 84 (1994) 323.

M. Ishimaru et al., Nucl. Instr. and Meth. B 166-167 (2000) 390.

Model

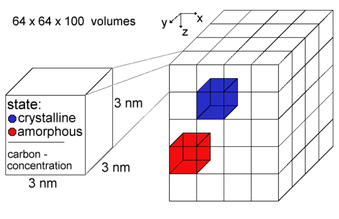
Model schematically displaying the formation of ordered lamellae with increasing dose.



- Supersaturation of C in $c-Si$
  → **Carbon induced** nucleation of spherical SiC_x-precipitates
- High interfacial energy between $3C-SiC$ and $c-Si$
  → **Amorphous** precipitates
- $20-30\,\%$ lower silicon density of $a-SiC_x$ compared to $c-Si$
  → **Lateral strain** (black arrows)
- Implantation range near surface
  → **Relaxation** of **vertical strain component**
- Reduction of the carbon supersaturation in $c-Si$
  → **Carbon diffusion** into amorphous volumina (white arrows)
- Remaining lateral strain
  → **Strain enhanced** lateral amorphisation
- Absence of crystalline neighbours (structural information)
  → **Stabilisation** of amorphous inclusions **against recrystallisation**

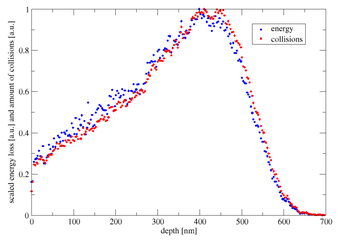
Simulation

Discretisation of the target



- divided into cells with a cube length of $3\,nm$
- periodic boundary conditions in x,y-direction

TRIM collision statstics



⇒ identical depth profiles for number of collisions per depth and nuclear stopping power

⇒ mean constant energy loss per collision

Simulation algorithm

The simulation algorithm consists of the following three parts looped s times corresponding to a dose $D = s/(64 \times 64 \times (3\,nm)^2)$:

loop

1. Amorphisation/Recrystallisation

- random numbers distributed according to the nuclear energy loss to determine the volume in which a collision occurs
- compute local probability for amorphisation:

$$p_{c\rightarrow a}(\vec{r}) = p_b + p_c c_C(\vec{r}) + \sum_{\text{amorphous neighbours}} \frac{p_s c_C(\vec{r'})}{(r-r')^2}$$

and recrystallisation:

$$p_{a\rightarrow c}(\vec{r}) = (1 - p_{c\rightarrow a}(\vec{r}))\left(1 - \frac{\sum_{direct\,neighbours}\delta(\vec{r'})}{6}\right),$$

$$\delta(\vec{r}) = \begin{cases} 1 & \text{if volume at position } \vec{r} \text{ is amorphous} \\ 0 & \text{otherwise} \end{cases}$$

- loop for the mean amount of hits by the ion

Three contributions to the amorphisation process controlled by:

- p_b normal 'ballistic' amorphisation
- p_c carbon induced amorphisation
- p_s stress enhanced amorphisation

2. Carbon incorporation

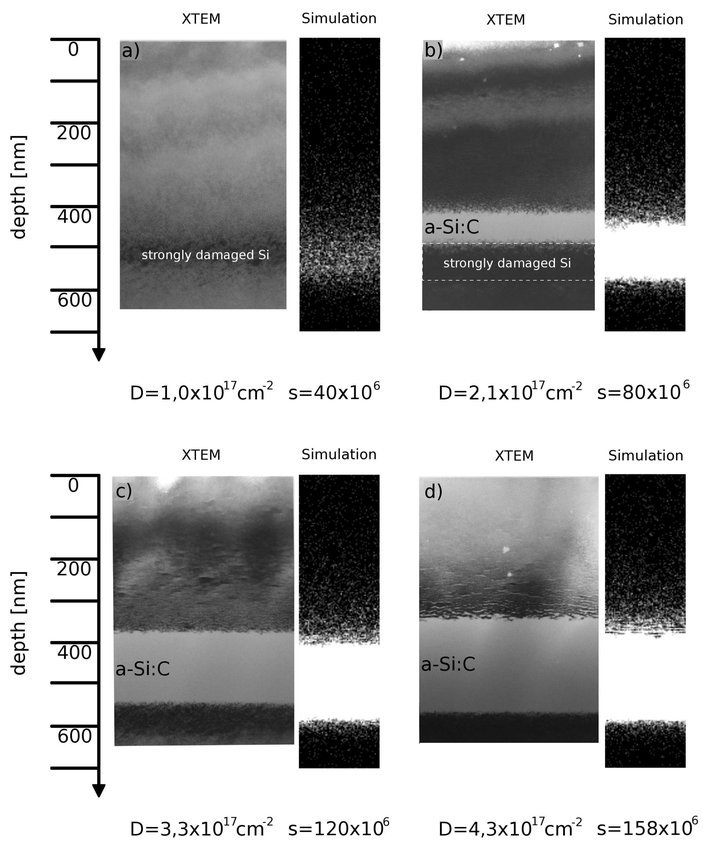
- random numbers distributed according to the implantation profile to determine the incorporation volume
- increase the amount of carbon atoms in that volume

3. Diffusion/Sputtering

Simulation parameters d_v, d_r and n control the diffusion and sputtering process.

- every d_v steps transfer of a fraction d_r of carbon atoms from crystalline volumina to an amorphous neighbour volume
- remove $3\,nm$ surface layer after n loops, shift remaining cells $3\,nm$ up and insert an empty, crystalline $3\,nm$ bottom layer

Comparison of experiment and simulation



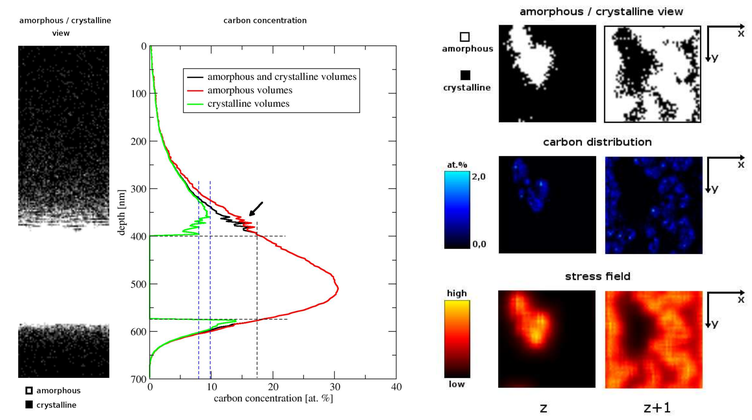
Simulation parameters:
$p_b = 0.01$, $p_c = 0.001 \times (3\,nm)^3$, $p_s = 0.0001 \times (3\,nm)^5$, $d_r = 0.05$, $d_v = 1 \times 10^6$.

Conclusion:

- Simulation in good agreement with experimentally observed formation and growth of the continuous amorphous layer
- Lamellar precipitates and their evolution at the upper a/c interface with increasing dose is reproduced

Simulation is able to model the whole depth region affected by the irradiation process

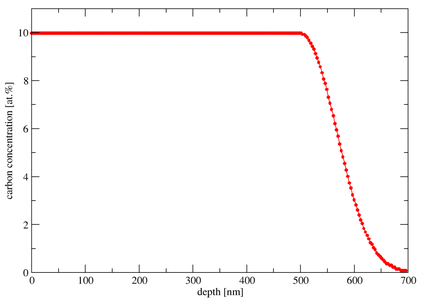
Structural/compositional information



- Fluctuation of the carbon concentration in the region of the lamellae
- Saturation limit of carbon in c-Si under given implantation conditions between 8 and 10 $at.\%$
- Complementarily arranged and alternating sequence of layers with high and low amount of amorphous regions
- Carbon accumulation in the amorphous phase

Recipe for thick films of ordered lamellae

Prerequisites:
Crystalline silicon target with a nearly constant carbon concentration at $10\,at.\%$ in a $500\,nm$ thick surface layer



Creation:

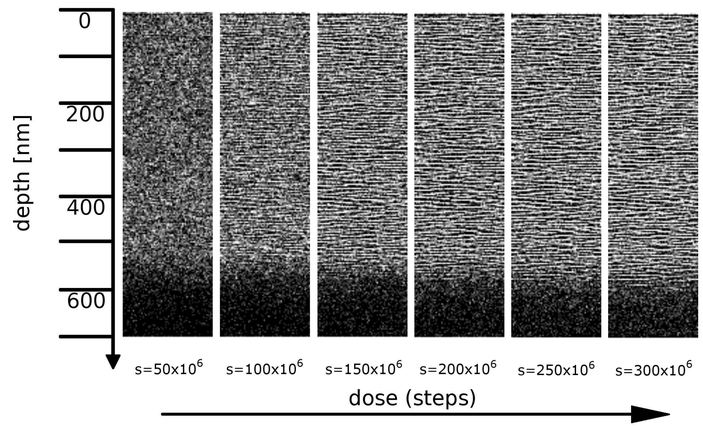
- Multiple energy (180-10 keV) $C^+ \rightarrow Si$ implantation
- $T_i = 500\,°\mathrm{C}$, to prevent amorphisation

Stirring up:

$2\,MeV\ C^+ \rightarrow Si$ irradiation step at $150\,°\mathrm{C}$

- This does not significantly change the carbon concentration in the top $500\,nm$
- Nearly constant nuclear energy loss in the top $700\,nm$ region

Result:



- Already ordered structures after 100×10^6 steps corresponding to a dose of $D = 2.7 \times 10^{17} cm^{-2}$
- More defined structures with increasing dose

Starting point for materials showing strong photoluminescence

Dihu Chen et al. Opt. Mater. 23 (2003) 65.

Conclusions

- Observation of selforganised nanometric precipitates by ion irradiation
- Model proposed describing the selforganisation process
- Model implemented in a Monte Carlo simulation code
- Modelling of the complete depth region affected by the irradiation process
- Simulation is able to reproduce entire amorphous phase formation
- Precipitation process gets traceable by simulation
- Detailed structural/compositional information available by simulation
- Recipe proposed for the formation of thick films of lamellar structure

Literature

F. Zirkelbach, M. Häberlen, J. K. N. Lindner, B. Stritzker. Comp. Mater. Sci. 33 (2005) 310.

F. Zirkelbach, M. Häberlen, J. K. N. Lindner, B. Stritzker. Nucl. Instr. and Meth. B 242 (2006) 679.