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Analysis of depth redistribution of implanted Fe near SiO_2/Si interface

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A B S T R A C T

We have studied diffusion and clustering processes of room-temperature (RT)-implanted Fe ions in a SiO_2/Si structure during annealing at 600 and 800 °C temperatures. The depth profile of implanted Fe was analyzed by Rutherford backscattering spectroscopy (RBS). In the previous study, we found that the hot-implanted Fe ions near the SiO_2/Si interface at high substrate temperatures of 600 and 800 °C were distributed significantly different from the result predicted in the TRIM simulation. We think that the diffusion phenomena during the ion implantation at such elevated temperatures are recognized to be strongly enhanced by ion-beam-irradiation effect. In this study, to simplify the diffusion phenomenon, we particularly treat thermal diffusion process of RT-Fe implantation around the SiO_2/Si interface in the post annealing at high temperatures. It is clearly seen that Fe atoms post-annealed at 800 °C are preferably gathered at a definitive depth in the SiO_2 layer around 15 nm distances from the interface. We finally compare the Fe depth distribution for hot-implanted samples to that for the post-annealed ones by RBS analysis quantitatively.

1. Introduction

We have developed a noble synthesis method of single-walled carbon nanotubes (SWCNTs) from metal clusters formed in SiO_2/Si substrates by hot-ion implantation technique so far [1,2]. Now, one is concerned with how to control the diameter and chirality of CNTs, since they define the electronic properties of band gap and electric conductivity [3]. In general, the diameter of CNTs is definitively responsible for that of metal clusters which behave as a catalytic seed of CNT synthesis. In order to fabricate CNTs with desired diameters, therefore, it is required to form uniform metal nanoparticles on/in the substrate. It is commonly known that most of metal ions implanted in oxides, such as SiO_2, MgO, and Al_2O_3, form clusters with a uniform size with several nm in diameter [4–6].

We have so far shown that the hot-implanted Fe ions in a SiO_2 layer had a significantly uniform size, and succeeded in synthesizing SWCNTs from the catalytic clusters using microwave-plasma-enhanced chemical vapor deposition (MPCVD), though we could not control the chirality of the SWCNTs. One believes that SWCNTs growth with a single chirality is generally responsible for the crystal orientation of the catalytic clusters. We then tried to form highly-oriented Fe clusters by Fe^+ hot-implantation just around the SiO_2/Si interface, because we expect both effects of clustering in the SiO_2 layer and epitaxial growth of Fe clusters on the single crystalline Si(001) substrate reflecting the orientation of the substrate.

We have recently studied growth of Fe clusters formed around a SiO_2/Si interface by hot-ion implantation at substrate temperatures of 300, 600, and 800 °C [2]. We characterized the implanted Fe using Rutherford backscattering spectroscopy (RBS) and transmission electron microscope (TEM). It was found in a TEM image for the 300 °C-implanted sample that tiny clusters with a mean diameter of 2.4 nm were formed in the SiO_2 layer. Besides, some of the clusters were found to be aligned near the SiO_2/Si interface. The depth profile of Fe obtained from the RBS analysis was not significantly different from the prediction from a Monte Carlo simulation by TRIM code. On the other hand, we observe nanoclusters with a mean diameter of 3.2 nm in the SiO_2 layer at a certain depth of ~10 nm apart from the interface for the 600 °C-implanted sample. We found significantly larger crystalline precipitations with size of 5–10 nm in diameter at the interface, which were assigned to be β-$FeSi_2$ nanoparticles by TEM analysis. Most of the implanted Fe atoms in the SiO_2 layer were segregated either on the SiO_2 surface or the SiO_2/Si interface when the implantation was performed at the substrate temperature of 800 °C.

It should be noted that the diffusion process during ion implantation at elevated temperatures is recognized to be significantly enhanced by ion-beam-irradiation effect. In the present study, we thus simply focus on the thermal diffusion process of RT-implanted Fe around the SiO_2/Si interface during post implantation annealing.

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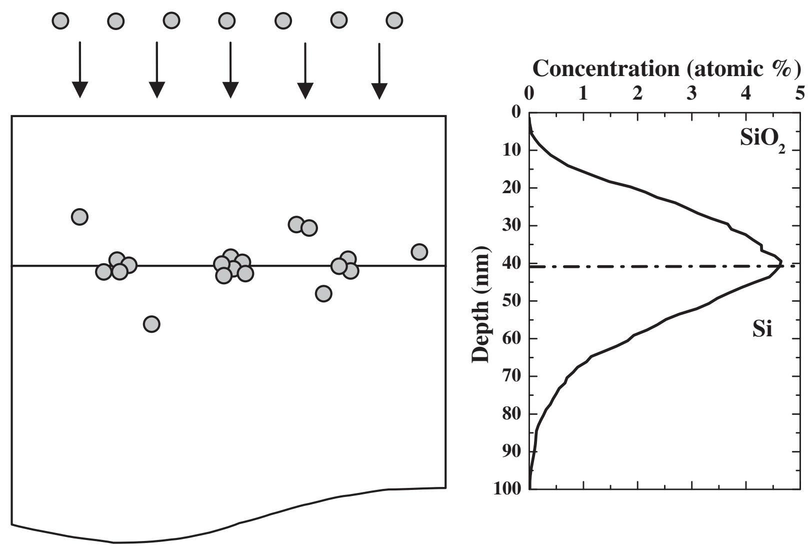

Fig. 1. A schematic image of the implantation along with the ion distribution simulated by the TRIM code assumed the parameters of the incident energy of 43 keV and the SiO_2 thickness of 40–45 nm.

2. Experiment

A SiO_2 layer was formed by dry thermal oxidation of Si(001) substrates at 900 °C for 2 h. The oxide thickness of the grown SiO_2 film was estimated to be about 40–45 nm by ellipsometry. We then optimized the incident energy of Fe ions by TRIM code in the SRIM2010 package [7], in order to implant the Fe ions as distributing around the SiO_2/Si interface. In this simulation, we assumed 2.21 g/cm^3 as the volume density of the SiO_2 layer [8]. As a result of the simulation, the optimized implantation energy was estimated to be 43 keV.

The ion implantation was performed using a medium-current ion implanter at Shonan–Hiratsuka Campus at Kanagawa University (SHC-KU). The implanter allows us to implant at wide substrate temperatures range from liquid nitrogen temperature to 1000 °C in a whole wafer with a 6-in. diameter uniformly by scanning accelerated ion beam with ~20 × 20 mm^2 size. The Fe ions are generated by sputtering a pure Fe repeller plate by Ar plasma ignited in an arc chamber. The maximum terminal voltage of the accelerator is 200 kV and the current density for the Fe ion with the energy of 43 keV typically is several hundred nA/cm^2. In the present study, the direct beam with 20 × 20 mm^2 size was irradiated on the sample of 8 × 8 mm^2 without beam scanning in order to reduce the time in the implantation. The Fe was implanted at room temperature (RT) in order to reduce the excess thermal diffusion enhanced by ion irradiation effect during the implantation. The Fe-implanted samples were annealed at 600 and 800 °C for 10, 30, and 60 min by an infrared radiation heater in vacuum of 10^{-5} Pa.

The depth profile of implanted Fe was analyzed by RBS using a pelletron accelerator placed at SHC-KU. In the present study, 1.8 MeV He^{++} ions were incident on the samples at a slightly tilted angle with respect to surface normal. The scattered primary ions with a grazing angle of 10° from the surface plain were detected by a Si surface barrier solid state detector. In order to obtain accurate random spectra, the sample was rotated around the surface normal during the measurements.

The observed spectra were analyzed by RBS simulation code by means of best-fitting the simulated spectrum to experimentally observed one by adjusting the fitting parameters defining the sample structures of elemental composition and layer thickness. The details of the simulation method can be found in some earlier papers [9–11].

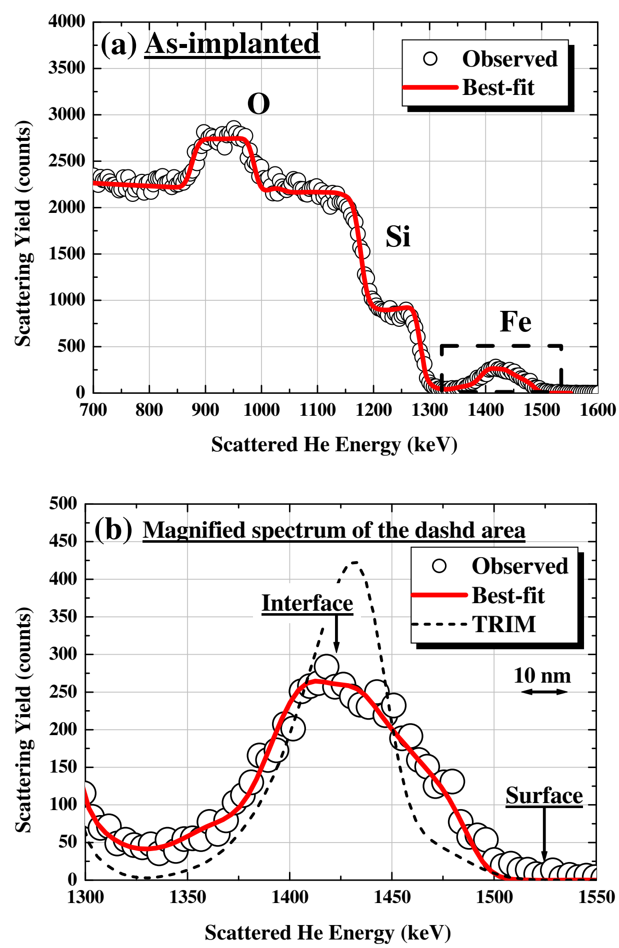

Fig. 2. Whole RBS spectrum (a) and magnified spectrum (b) observed for the as-implanted sample at RT with ion dose of 1 × 10^{16} ions/cm^2. Circles and a solid curve indicate experimentally observed spectrum and the best-fitted one, respectively. A dotted curve corresponds to the spectrum assumed the depth profile obtained from the TRIM simulation.