Paper

Angular Distribution of Sputtered Particles in Shave-off Section Processing with SDTrimSP

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The angular distribution of secondary ions is one of the essential elements for the development of three-dimensional (3D) shave-off SIMS. The magnification lens system in the 3D shave-off SIMS was designed and assembled based on the detection position of the secondary ion, the emission angle of the secondary ions defined the detection position on the detector. However, shave-off condition of high incidence energy (30 keV) and high angle of incidence (87 degrees), we simulated the angular distribution of sputtered particles using the SDTrimSP program and compared the results with the previous shave-off experimental data. Even unusual the shave-off beam, the SDTrimSP simulation results showed a good agreement and a similar tendency with the experimental data. SDTrimSP simulation is expected to be useful in obtaining the sputtered particle information for development and instrumentation of the 3D shave-off SIMS.

1. Introduction

The shave-off method has been studied for achieving highly precise depth profiling using nano-beam secondary ion mass spectrometry (SIMS) [1, 2] with a protection film [3], flood gun system [4], and multilane profiling [5]. In particular, multilane profiling enables one to acquire the lateral information, and it has developed for the data acquisition of shave-off SIMS in two-dimensions (2D) with good resolution [6]. As miniaturization progresses and development in the field of materials and life science, the 3D analysis of material structure is required with excellent spatial resolution and sensitivity for chemical information. Recently, our group has studied and simulated new three-dimensional (3D) shave-off SIMS method, which added the depth information to the multilane profiling of the 2D shave-off method [7].

In the 3D shave-off system, we introduced the cylindrical lens system, which magnifies the depth of the locational information of secondary ions from a few micrometers to some millimeters unit. The magnified sample depth information appears in the Y-axis of a detector using an elemental analysis (mass information m/z, X-axis of the detector).

In our previous report [8], the sample depth locational information that appeared on the detector depended on the initial energy and the angular distribution of secondary ions. In particular, the angle distribution critically affected the detection position of the secondary ions, and it is the most critical and essential element for the development of the 3D shave-off method development.

In this study, we simulated the angular distribution of sputtered particles using the SDTrimSP code under the shave-off condition. The SDTrimSP code, one of the Monte Carlo simulations of the binary collision process, is known for its good agreement with experimental data of angular distribution better than other simulation programs under conventional scanning conditions [9]. However, the simulation program has no references for shave-off conditions, which is high energy (over 20 keV), and has a high angle of incidence (over 80 degrees) of gallium focused ions. Therefore, in order to verify that the SDTrimSP simulation results are in good agreement
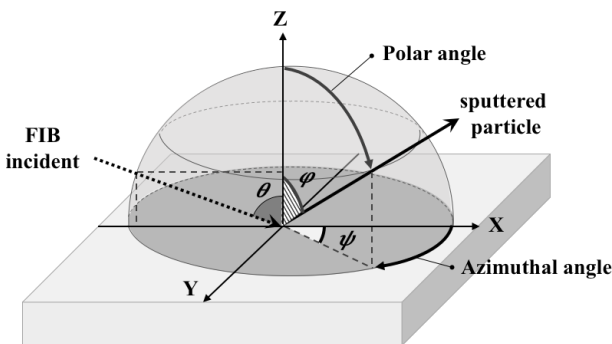
with the experimental data even under the shave-off condition, we simulated and compared the angular distribution of sputtered particles based on the previous experimental data [10].

2. Method

All of the simulations for the angular distribution were performed with the SDTrimSP program (version 5.07) [11]. The input parameter of the simulation followed each existing experimental condition, and the surface binding energy of the target was 3.79 eV (Au) and 4.72 eV (Si), which were tabulated by the SDTrimSP program. The simulation used detailed calculations using 50,000 of Au particles and 605,000 of Si particles among the sputtered particles from 100,000 incidents of Ga^+ ions. When the primary ion beam is irradiated on the sample surface with an incident angle (θ), the angular distribution is indicated by the polar angle (φ) and azimuthal angle (ψ) of the sputtered particle (Figure 1).
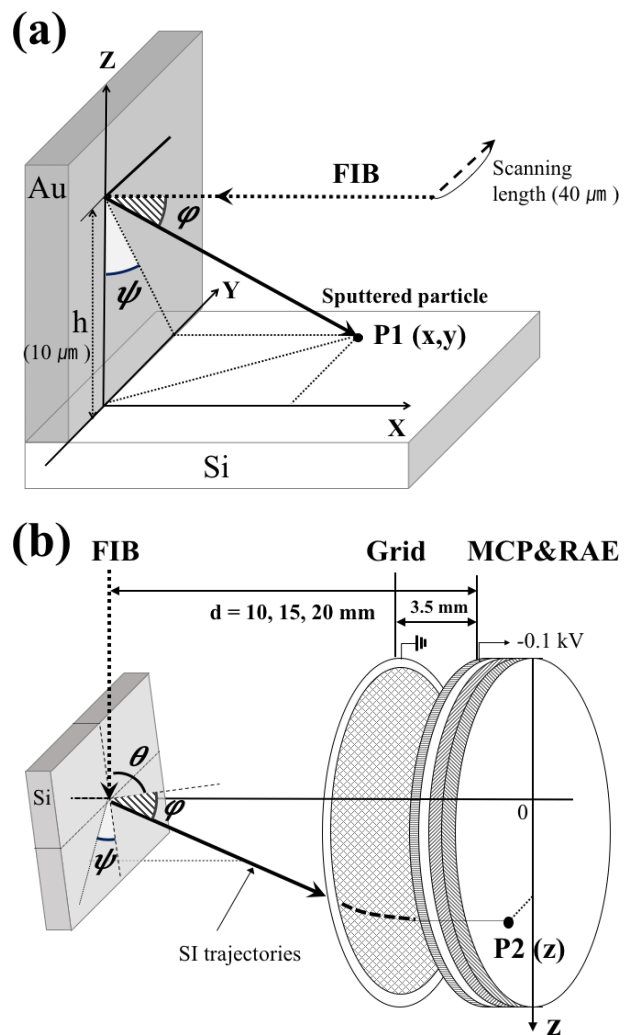
First, as the condition of high incident energy, 30 keV of Ga^+ ions irradiated a gold film at a normal incident angle ($\theta = 0°$). The angular distribution of the gold particles that were sputtered from beam scanning of 40 µm along Y-axis was measured from the deposit position on the silicon substrate. The silicon substrate is located vertically at the height (h) of 10 µm, and the deposit position (P1) is calculated by using the simulation results of the polar angle and azimuthal angle as shown in Figure 2(a).

The second investigation was conducted at a high angle of incidence. A silicon wafer is bombarded by Ga^+ ions of 20 keV at 87 degrees. Figure 2(b) depicts the experimental system for measuring the angular

distribution of secondary ions with a high angle of incidence. The secondary ions, which had energy under 10 eV, were measured by a position sensitive detector. The detector system is composed of a grounded mesh grid, microchannel plates (MCP), and a resistive anode encoder (RAE), all of which are located parallel to the FIB (focused ion beam) from a distance (d). The angular distribution of silicon sputtered particles was calculated as the arrival position (P2) on the detector using the SDTrimSP and SIMION programs. The angular distribution was examined by changing the distance (d) between the sample and the first layer of MCP as 10 mm, 15 mm, and 20 mm.
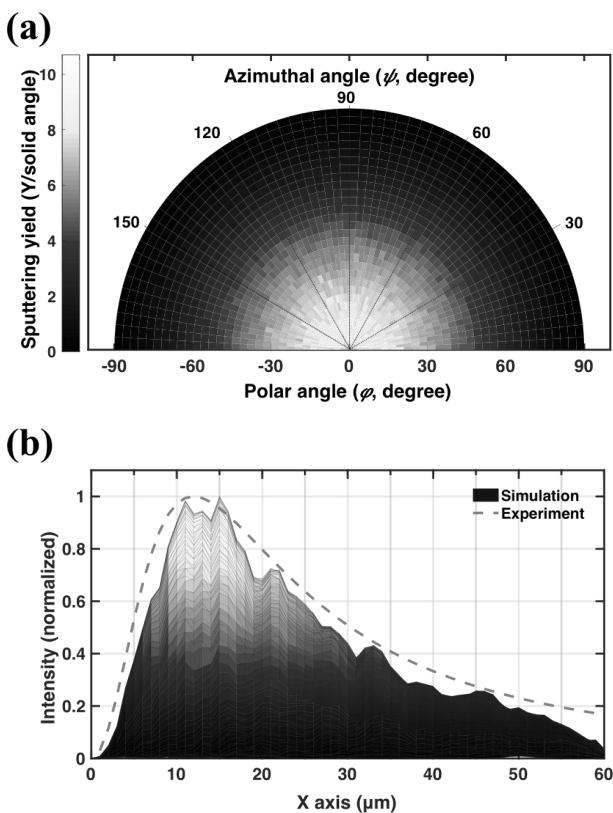
(a)

(b)

Fig. 2 Schematic illustration of the shave-off experimental conditions for a high incident energy (20keV) erosion of a gold film at a normal incident angle (a), and a high incident angle (θ = 87 degrees) erosion of silicon substrate (b).

Fig. 1 Geometry for the sputtered particles, where θ denotes the angle of incidence, φ is the polar angle and ψ is the azimuthal angle.

3. Results and Discussion

3.1 High incident energy

In the first example for high incident energy, the gold film was irradiated by 30 keV of Ga⁺ ions at the normal incident angle. Figure 3(a) shows the simulation result for the angular distribution of the sputtering yield per solid angle. The sputtered gold particles appeared to be distributed symmetrically at azimuthal angles (0–180°), whereas at the polar angles below about 30 degrees showed high emission distribution.

For comparison to existing experimental data [10], we calculated the deposit positions of gold sputtered particles using the simulation results. Figure 3(b) shows the simulation result and the experimental data of the side view along the X-Z plane in Figure 2(a). In the experiment, the thickness of the deposit film on Si substrate was measured by a confocal laser scanning microscope (VK-X250, KEYENCE), and the cross-section shape was indicated as a dashed line in Figure 3(b). In the simulation, the sputtered gold particles

deposited on the Si substrate was plotted as a histogram count. The simulation result shows good agreement with the experimental data. The emission angle calculated from the peak top of the graph (15 μm) was about 33.7 degrees ($\tan^{-1}(10/15)$). The emission angle was also in accordance with the results of the polar angular distribution in Figure 3(a).

3.2 High angle of incidence

For the condition of the high angle of incidence, 20 keV of Ga⁺ ions were irradiated on the Si with 87 degrees angle of incidence. The simulation results of the angular distribution for the sputtered Si particles are shown in Figure 4. Most of the sputtered particles were toward the polar angle of 0–30 degrees and the azimuthal angle of 0–90 degrees.

In the experiment, the electric field is generated between the grounded mesh and the first layer of the MCP (-0.10 kV), and it was expected to affect the trajectories of secondary ions. Therefore, the angular distribution was calculated including the electric field effects by using the SIMION program. The emission information (polar angles, azimuthal angles, and energy) were obtained by an SDTrimSP simulation and, based on this, the final arrival position (P2 in Figure 2(b)) on the detector was calculated using the SIMION program. Figure 5 shows comparison of the simulation results and experimental results when the distance between the sample and detector is d = 10 mm (a), 15 mm (b), and 20 mm (c), respectively. The particle counts of each result were normalized for comparison, and the sample position
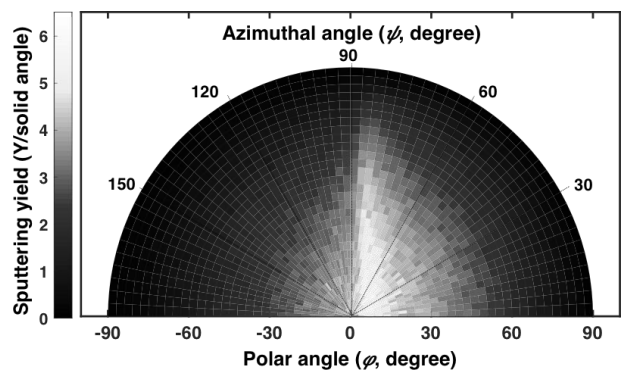
(a)

(b)

Fig. 3 (a) Contour plot of the angular distribution of the sputtered Au particles. The color bar indicates the sputtering yield per solid angle for Au atoms (b) Comparison between simulation results and experimental data of deposit particles.

Fig. 4 Simulation result of the angular distribution of the sputtered Si particles for 20 keV of Ga⁺ ion irradiation with a high incidence angle of 87°. The color bar represents the sputtering yield per solid angle.
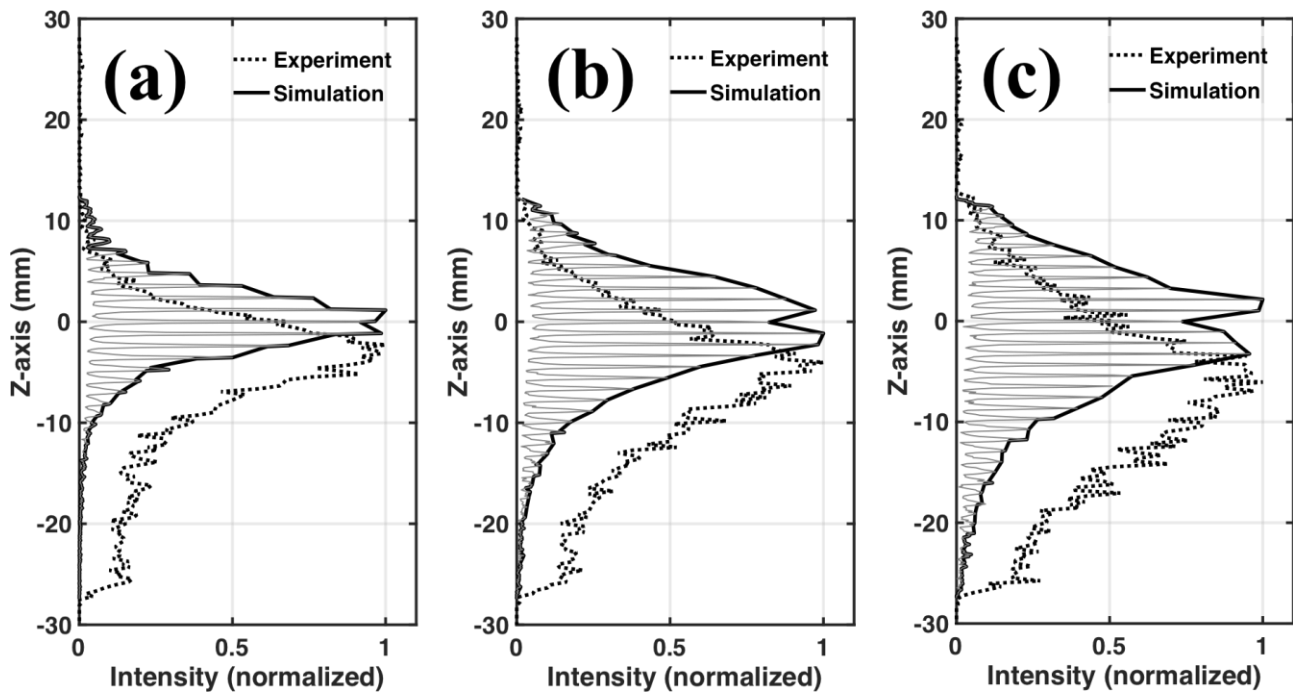
Fig. 5 The number of normalized counts along the Z-axis of the detector when the distance (d) between the sample and the detector are 10 mm (a), 15 mm (b), and 20 mm (c).

of the Z-axis was set at zero (0 mm). All three simulation results were distributed above the experimental data. The experimental results show that the emission direction of secondary ions is going downward (a negative value of the Z-axis). However, in the simulation, most of the sputtered silicon particles having energy under 10 eV were emitted in a vertical direction from the FIB (0 mm of the Z-axis). The peak of the simulation distribution graph shows split ends, this is supposed to occur due to the pass of the mesh grid. As the length (d) increases, the area of distribution widens in both simulations and experiments, the difference between the simulation results and experimental data also increases. The angular distribution of simulations shows a difference of about 3 mm (d = 10 mm), 5 mm (d = 15 mm), and 7 mm (d = 20 mm) from the experimental data. The difference was represented by the average emission angle of about 18 degrees (the average angles of $\tan^{-1}(3/10)$, $\tan^{-1}(5/15)$, and $\tan^{-1}(7/20)$). The simulation results had smaller emission angles than the experimental results, but they show quite a similar shape of distribution to the experimental results for the angular distribution of sputtered particles.

4. Conclusion

The investigation of the angular distribution of sputtered particles is an important and basic parameter to calculate the secondary ion trajectories for development of the 3D shave-off SIMS apparatus. At the shave-off conditions, the angular distribution of sputtered particles was simulated using the SDTrimSP simulation software, and the results were compared to existing experimental data.

First, we simulated the angular distribution of sputtered gold particles for a high incident energy with 30 keV of Ga$^+$ ions irradiated at the normal angle of incidence (0 degree). The angular distribution was measured by calculating the deposit position, and the simulation result shows good agreement with the experimental data.

For the simulation under the condition of a high angle of incidence, the silicon is irradiated with 20 keV of Ga$^+$ ions incident at 87 degrees. The angular distribution of sputtered silicon particles showed a polar angle of 0–30 degrees, and the sputtered particles having energy under 10 eV exhibited a perpendicular direction to the FIB. However, the experimental results showed that the

angular distribution of secondary ions is directed downward with a difference of about 18 degrees with the simulation results. Although the simulation results are not completely in accordance with the direction of the shave-off experimental data, they showed a similar tendency of distribution shape with the experimental data. The SDTrimSP simulation is expected to help obtain the angular distribution of sputtered particles under shave-off conditions. Moreover, it will be able to provide the basic parameter for calculating the secondary ion trajectories along the lens system and detector in the development of 3D shave-off SIMS.

5. Acknowledgement

6. References

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Discussion and Q&A with Reviewers

Reviewer #1 (private)

[Q1-1] Introduction

Please describe the advantages for material analysis by improvement 2D to 3D.

[A1-1]

As suggested by reviewer, I have added description

The sentence now reads;

" In particular, multilane profiling enables one to acquire the lateral information, and it has developed for the data acquisition of shave-off SIMS in two-dimensions (2D) with good resolution [6]. As miniaturization progresses and development in the field of materials and life science, the 3D analysis of material structure is required with excellent spatial resolution and sensitivity chemical information. Recently, our group has studied and simulated new three-dimensional (3D) shave-off SIMS method, which added the depth information to the multilane profiling of the 2D shave-off method [7]. "

[Q1-2] Method

"The input parameter of the simulation followed each existing experimental condition, and the surface binding energy of target was 3.79 eV (Au) and 4.72 eV (Si) which were tabulated by the SDTrimSP program."

→Author should describe the reason for using two kinds of samples (Au and Si). The reviewer guesses that the reason for using Au was convenient for measuring deposited films.

[A1-2]

This paper compares the SDTrimSP with previous (existing) experiments results to see whether the SDTrimSP has a good agreement for the shave-off method. Therefore, there is no reason for choosing the kinds of samples.

[Q1-3] Method

"The simulation used detailed calculation with the sputtered particles (50,000 of Au particles and 605,000 of Si particles) from 10,000 of incident Ga$^+$ ions."

→Is the sputtering rate of Si so high?

[A1-3]

50,000 of Au particles and 605,000 of Si particles are just number of particles extracted and used for simulation. It has nothing to do with the sputtering yield. So I changed the sentence.

The sentence now reads;

"The simulation used detailed calculations using 50,000 of Au particles and 605,000 of Si particles among the sputtered particles from 100,000 incidents of Ga$^+$ ions."

[Q1-4] Method

"The secondary ions which had energy under 10 eV were measured by position sensitive detector."

→Please describe the reason and method for detecting the secondary ions under 10 eV.

[A1-4]

In the reference experiments, the author had investigated from 1 eV to 20 eV, but over the 10 eV, he had not seen significant difference of the peak positions, so he did experiment with the secondary ions which had 10 eV.

[Q1-5] Results and discussion, High Incident energy

"In the experiment, thickness of the deposit film on the Si substrate were measured by a confocal laser scanning microscope"

→Author should describe the model number of the microscope.

[A1-5]

This experiment is not my results, it is just reference, so I didn't write details, but I added model number of the microscope from the reference.

The sentence now reads;

"In the experiment, the thickness of the deposit film on the Si substrate was measured by a confocal laser scanning microscope (VK-X250, KEYENCE),"

[Q1-6]Result and discussion, High angle of incidence

"The peak of the simulation distribution graph shows split ends, it is supposed to occur due to the pass of the mesh grid."

→Reviewer think that the shadow area on the detector

due to the grid of the mesh become smaller as increasing the distance between sample and the detector. The split of peak top become smaller depending on the distance. However, the result of simulation does not agree.

[A1-6]

When the distance between the sample and the detector is increasing, the distance between the mesh and the detector is the same (keep). Therefore, the shadow area of the detector does not change. As the distance increases, the distribution of the ions becomes wider, so that the split of the peak top is expected to increase.

I changed the Fig. 2(b) to include the distance between the mesh and the MCP (3.5 mm).

[Q1-7] Conclusion

"However, the experimental results showed that the angular distribution of secondary ions is directed to downward with a difference about 18 degrees with simulation results. Although the simulation results are not completely in accordance with shave-off experimental data, they showed an acceptable similar tendency for the experimental data."

→Please comment about the reason for a difference of 18 degrees. Reviewer thinks that 18 degrees is a big difference, but what is the reason for judging that it is acceptable?

[A1-7]

The peak of distribution showed difference of 18 degrees, but the simulation results showed the similar shape of angular distribution with experiment. So I wrote the acceptable tendency. But I had rewrote the sentence to clarify

The sentence now reads;

(**3.Results and discussion- High angle of incidence**)

"The simulation results had smaller emission angles than the experimental results, but they show quite a similar shape of distribution to the experimental results for the angular distribution of sputtered particles."

(**4.Conclusion**)

"However, the experimental results showed that the angular distribution of secondary ions is directed downward with a difference of about 18 degrees with the

simulation results. Although the simulation results are not completely in accordance with the direction of the shave-off experimental data, they showed a similar tendency of distribution shape with the experimental data."

Reviewer #2 Akio Takano (Toyama)

Peer review content private.