Angular distributions of sputtered silicon at grazing gallium ion beam incidence

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Abstract

Angular distributions of silicon atoms sputtered by gallium ions at grazing incidence were investigated experimentally and by simulation. The energies and the ion beam fluence studied are typical for using focused ion beam techniques for silicon micro-structuring. The angular distributions of the sputtered atoms at grazing ion beam incidence loose their cylindrical symmetry around the target surface normal due to the anisotropy of single collisions and of the collision cascades initiated by the energetic ions. The angular distributions were studied in this work both experimentally and using the simulation methods of Monte-Carlo and of Molecular Dynamics. To study the influence of the surface structure on the angular distributions of the sputtered atoms, different arrangements of silicon atoms on the surface of the targets were tested in simulations using Molecular Dynamics. Monte-Carlo simulations based on the theory of binary collisions were used to complement the experimental results on the angular distributions of sputtered silicon and the final results are presented as an analytical model.

Introduction

There are several applications of ion beam sputtering in which the angular dependence of the total sputtering yield and the angular distribution of the sputtered particles are essential. For example, the inclination of the walls of the trenches etched in silicon by the focused ion beam (FIB) sputtering technique is determined by an interplay of the angular dependence of the sputtering yield and by the angular distribution of the sputtered atoms. Therefore, for an adequate simulation of the geometry of the silicon structures produced using the FIB sputtering technique, a profound knowledge on the angular distributions of the sputtered material is necessary. Motivated by this necessity, angular distributions of the sputtered silicon irradiated by the gallium ion beam were investigated in this work. The energy of the gallium ion beam in this work was 30 keV and the angles of incidence between 60º to 80º from the normal to the silicon surface. The incidence angle of 80º roughly corresponds to the maximum of the sputtering yield. Simulations based on the Monte-Carlo technique [1]-[5] show that the sputtering yield for silicon sputtered with a gallium ion beam of 30 keV at a tilt angle of 80º is about a factor of 7 larger than at normal incidence. A very strong anisotropy of the angular distributions with distinct ridges in forward direction was reported in the literature in case of metallic targets sputtered with light ions [6]. In contrast, the silicon target studied in this work has a completely different kind of inter-atomic binding in comparison to metals. Since the case of silicon sputtering with gallium ions has a large application potential in the FIB technique, this topic was already addressed in the literature [7]-[9]. Nevertheless, the published data are not sufficient to build a complete model that is needed to simulate the FIB processing. Therefore, the task of this work is to study the angular distributions of the sputtered silicon atoms, especially at grazing incidence of gallium ion beam. For this purpose, angular distributions of the sputtered atoms were measured using the collector technique. Furthermore, numerical simulations of sputtering were performed to complement the experimental data for the angular range not well accessible in the measurement. Finally, an analytical model for the angular distributions of the sputtered silicon suitable for the simulation of the FIB processing was proposed.

Methods

Two experimental setups were realized in a vacuum chamber of a FIB system to measure angular distributions of sputtered silicon: the first one for the analysis of the sputtered material using the Rutherford Backscattering Spectroscopy (RBS) and the second one for the analysis using time-of-flight (TOF) SIMS. In the first setup, the collector sample was aligned parallel to the target surface and the distance between the target and the collector was 5.5 mm. Gallium ions with a kinetic energy of 30 keV bombarded silicon under angles of incidence between 60° and 80°. The collector was a piece of a silicon wafer with a 3 µm polymer coating on it. The coating was used for collecting the sputtered silicon atoms, and the silicon wafer at the bottom served for mechanical stability and also as heat sink during the RBS analysis. The RBS measurements were performed at the University of Minsk (Belarus) with a He ion beam of 1 MeV energy and a spatial resolution of 0.3 mm. The specified spatial resolution of the RBS measurements results in an angular resolution of about 6° near the normal to the silicon surface direction.

The second experimental setup differs from the first one by a different collector. In the second setup, plane Kapton polyimide foil was used as collector. The collector foil was tilted so that the angle between the target plane and the collector plane was 58º. In this geometrical arrangement, for the particles sputtered in the forward direction in respect to the ion beam the distance from the target to the collector is smaller than for the particles sputtered in the backward direction. The Kapton collector was analyzed using the time-of-flight secondary ion mass spectrometer of type TOF.SIMS 5. Bismuth ions with an energy of 25 keV were used as primary ions in the SIMS analysis.

The numerical simulations of the ion beam sputtering were performed using two methods. First, the Monte-Carlo method based on the binary collision approximation with the ZBL-inter-atomic potential [1] was used. In this work, we used the implementation of this method in the computer code MCSIM [5] developed at the Fraunhofer Institute of Integrated Systems and Device Technology. Since the ZBL-potential is a purely repulsive one and does not include the attractive forces at large inter-atomic distances, the surface is treated in MCSIM as a potential barrier with a height equal to the surface binding energy. The standard value of the surface binding energy for silicon is 4.7 eV [1]. An important improvement that was made in MCSIM in comparison to the TRIM program [1]-[3] was the introduction of the local electronic stopping according to Oen-Robinson [10] for low energies of silicon atoms and a continuous transition to the ZBL [1] electronic stopping power in the energy range 0 to 3 keV. The second simulation method used here is the method of Molecular Dynamics (MD) [11]. The implementation of the MD method in the computer code XMD-2.5.34-1 [12], [13] was used. The method of MD captures rigorously all the possible interactions between the incident ion and the target atoms involved, therefore more accurate results for sputtering simulation are expected. The XMD program contains several implementations of the inter-atomic potentials. For this work, a combination of the Tersoff-potential [14] with the ZBL-potential [1] as described by Belko et al. [15] was implemented in XMD and was used in MD simulations. A rectangular block with a width and length of 92 Å and a thickness of 38 Å containing 16184 silicon atoms was used in MD simulations. Since silicon is amorphized already at a relatively low fluence of gallium ions of about 10^{15} cm^{-2} and typical fluences in the FIB processing exceed 10^{17} cm^{-2}, we modeled an amorphous sample of silicon in a preliminary XMD simulation. First, a high temperature over the melting point of silicon was simulated using XMD, then a rapid cooling down of the sample was modeled using the quenching command of the XMD program. In the result, the atomic structure of the silicon sample was as it is expected for an amorphous state. The initial conditions for the silicon atoms were set by defining the initial temperature of the sample to 300 K. A moving gallium atom with a velocity of $2.88\cdot10^5$ m/s corresponding to an energy of gallium atoms of 30 keV was assumed in the simulation. The direction of the ion impact corresponded to the experimental setup and the initial positions of gallium atoms were randomly distributed in a square with a side length of 5.43 Å equal to the lattice constant of silicon crystal and which was oriented parallel to the sample surface. The MD simulation consisted of two stages. In the first stage which lasted 250 fs, the penetration of the gallium ion into the silicon block was simulated. The duration of the first stage was long enough that the gallium ion left the simulation sample. In the second stage which was 500 fs long, the relaxation of the atomic collision cascade inside of the simulation sample was modeled. The duration of the second stage was long enough that the vast majority of the sputtered particles were already emitted. Periodic boundary conditions were assumed at the side walls of the simulation sample and the top and the bottom of the sample were treated as free surfaces. The particles emitted from the bottom surface were simulated to the end of the second stage in order to control the total energy of the system which was constant due to the adiabatic assumption. The silicon atoms emitted from the top surface of the sample were marked as the sputtered particles and their positions and angle of emission were recorded into a file.

Results and Discussion

First, the two-dimensional distribution of the sputtered silicon under gallium beam irradiation is presented. Figure 1a shows the 2D angular distribution of sputtered silicon for 60º ion beam tilt obtained experimentally using TOF-SIMS and Figure 1b shows the same distribution simulated by MCSIM. According to the symmetry of the experimental setup, the distribution should be left-to-right symmetrical. The ion beam projection onto the collector plane shown in the figure is along the positive direction of the y_{2D}-axis. The top-down asymmetry of the distribution is partly due to geometrical arrangement of the collector relative to the target as described in the previous section and partly due to asymmetry of the forward-backward emission directions for a tilted ion beam alignment. The simulation results of MCSIM well reproduce the essential features of the measured two-dimensional distribution.

Next, the experimental results obtained using the RBS measurements are presented. The statistical uncertainty of the RBS measurement estimated as the standard deviation of the Poisson distribution of the number of the particle counts near the maximum of the angular distribution amounts to 1.7%. Figure 2 shows the distributions of the sputtered silicon measured on a planar collector sample which has been aligned parallel to the silicon sample. The locations of the RBS measurements on the collector were taken on a line along the projection of the beam onto the collector surface. Figure 2a shows the distribution of the sputtered silicon atoms for 70º and Figure 2b for 80º tilt angle. The zero angle in Figure 2 is for the emission direction normal to the sample surface, positive angles of 20º and 10º correspond to an emission direction normal to the ion beam tilted by 70º and 80º, respectively. For comparison, Monte-Carlo simulation results are shown. It should be noted that the overall shape of the angular distributions of the sputtered silicon is well reproduced by MCSIM, but there are also differences seen between the measurements and simulation. A distinct local peak in the angular distribution of the sputtered silicon at about 24º in Figure 2a is observed in MCSIM simulation, but cannot be seen in the measurement. The peak in the simulation comes from a direct knock-out of target atoms by the gallium ions. For 80º ion beam tilt, a peak due to the direct knock-out is observed in Figure 2b at about 13º both in the measurement and in the simulation. On the other hand, the absolute maximum of the sputtered atom emission in the measurements is at zero angle in Figures 2a and 2b, this means in direction normal to the sample surface. This comparison indicates that MCSIM overestimates the intensity of the sputtering peak associated with the direct knock-out of the target atoms.

To elucidate possible reasons for the overestimation of the direct knock-out maximum in MCSIM, we performed simulations with the MD method using the XMD program. Since XMD needs much more computing effort, we could not perform a direct comparison of XMD results with experiments on angular distributions. Instead, we analyzed how angular distributions of the sputtered atoms change, if different assumptions about the atomic structure of the target are taken. To ensure a better statistical accuracy of the XMD simulations, one-dimensional angular distributions of the sputtered atoms were considered. These distributions have been obtained from 2D angular distributions by integration over the emission directions that are obtained by a rotation around the projection of the ion beam onto the target surface.

First, the silicon target was approximated by a single atom in the XMD simulations and the angular distribution of the target atoms emitted after the irradiation with the gallium ion beam was investigated. This simulation should demonstrate that the effect of the direct knock-out of the target atoms can also be described using the MD method. The single-atomic target was used to observe what happens if inter-atomic interactions in the target are completely switched-off. In this case, only atomic scattering due to direct knock-out of the target atoms happens and therefore a single peak of the sputtered atoms appears in direction close to the normal to the ion beam direction as shown in Figure 3. This simulation also shows that for an adequate simulation of the interaction of the fast incident ions with relatively slow moving target atoms very short time steps must be used. For the case of 30 keV gallium ions, the shape of the sputtering peak does not change essentially, if a time step of $1 \cdot 10^{-16}$ s or less is used in XMD simulation. To ensure a high accuracy of angular distributions, a time step of $1 \cdot 10^{-16}$ s was used in all the following XMD simulations.

After all the relevant XMD simulation parameters were tested and verified using the single-atom target, XMD simulations for silicon samples with different atomic structure of the surface were performed. A planar surface, a surface containing 25% of isolated atoms, a surface containing 27% of atoms arranged in atomic rows, and a mono-layer step were considered. The total number of the primary gallium ions used in the simulation of each distribution shown as a line in Figure 4 was 1000. If the inter-atomic interactions in a solid target are considered in XMD, the angular distribution of the sputtered atoms spreads over all the possible emission directions as shown in Figure 4. The maximum of the emission of the silicon atoms in the XMD simulations is also as in the MCSIM simulations not in direction of the target normal but it is shifted to the forward direction by 10° to 20°. The angular distributions of the sputtered silicon simulated for differently structured surfaces show that the variations of the surface structure of the silicon target changes the total sputtering yield but has only a small impact on the angular distributions. It should be noted that the total sputtering yield obtained from XMD simulations agrees well with experimental measurements. The experimental sputtering yield was obtained from the measurements of the sputtering craters by the scanning electron beam in the FIB machine.

Table I presents the results on sputtering yield of silicon for gallium ion sputtering at grazing beam incidence of 80°. The simulated sputtering yield changes between 19.6 to 23.8 depending on the atomic structure of the surface and the experimental value of 19 is close to this range. It is somewhat unexpected but the highest sputtering yield was obtained for the flat surface. The sputtering yield diminishes if the atomic coverage of the surface layer diminishes and it is the lowest for a surface with a mono-layer step. The gallium ion beam was oriented 45° to the mono-layer step line when looking from the top onto the silicon target and the side wall of the step was exposed to the ion beam.

Since the MCSIM code reproduces the essential features of the angular distributions of silicon at different tilt angles, we applied MCSIM for the simulation of the angular distributions of silicon for different angles of gallium irradiation and summarized the results in an analytical model. The analytical model for the angular distribution of the sputtered atoms $Y_S(\vartheta,\varphi)$ at tilted ion beam impact reads:

$$Y_S(\vartheta,\varphi) = \begin{cases} G_1(\vartheta)\cos^n(\varphi) & for \quad \vartheta \le \vartheta_{PKA} \\ G_2(\vartheta)\cos^n(\varphi) & for \quad \vartheta \ge \vartheta_{PKA} \end{cases},$$

where $G_1(\vartheta)$ and $G_2(\vartheta)$ are two Gauss functions each defined by a mean value μ, a standard deviation σ, a pre-factor A, and a back-ground correction y that is added to the standard

Gaussian distribution; ϑ is the emission angle in the forward-backward direction in respect to the projection of the ion beam onto the target surface (positive values for the forward direction, negative for the backward direction); φ is the azimuthal angle that has its zero value in the plane which contains the ion beam and the surface normal, it ranges from -90° to +90° indicating the left-hand side emission direction by negative values and the right-hand side one by positive values; ϑ_{PKA} indicates the angular position of the sputtering peaks due to a direct single-collision knock-out of the target atoms. For example, the numerical parameters of the analytical formula for the case of silicon sputtering with a 30 keV gallium ion beam tilted by 70° are as follows: μ_1=13.8°, σ_1=64.1º, A_1=1121.8, y_1=-1.85, μ_2=3.68°, σ_2=64.8°, A_2=1591.3, y_2=-4.04, n=1.12, and the parameter ϑ_{PKA} is approximated by ϑ_{PKA} = 25.38° – 16.15°·φ/90°. A comparison of the analytical formula with the results of the numerical simulation using MCSIM is presented in Figure 5. The analytical formula describes the numerically simulated distributions very well except for the fact that the small local peak due to the direct knock-out of the silicon atoms which appears in MCSIM simulations is represented by a small discontinuity in the analytical modeling. The justification for this approach is the suppression of this peak in the XMD simulations, and also the absence of the distinct peak in the measurements at tilt angles smaller than 80°.

Conclusion

The angular distributions of the sputtered silicon during the gallium ion beam irradiation of silicon at tilted ion beam incidence were investigated. In contrast to the symmetrical angular distributions observed at normal ion beam incidence, asymmetrical distributions result for tilted ion beam impact. The Monte-Carlo simulation based on the binary collision approximation implemented in our computer code MCSIM was able to reproduce the essential features of the measured angular distributions of the sputtered atoms. On the other hand, MCSIM somewhat overestimates the intensity of the peak in the angular distributions that is associated with a direct knock-out of the target atoms and underestimates the sputtering in direction close to the normal of the target at larger ion beam tilt angles. Simulations based on the method of molecular dynamics were used in an attempt to improve the qualitative agreement with the measurements and for a study of the effects of the surface structure. Although the XMD simulations show a strong randomization of the emission of the sputtered particles due to inter-atomic interactions in the target, the maximum of sputter emission at grazing ion beam incidence remains in the direction nearly perpendicular to the ion beam, not to the surface normal as it is observed in the measurements. An analytical model is suggested to describe the asymmetrical angular distributions of the sputtered material at the tilted ion beam impact.

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Table I: Total sputtering yield for silicon irradiated by a 80° tilted gallium ion beam with an energy of 30 keV

XMD Simulation				Experiment
Flat surface	25% atomic coverage	27% atomic rows	A mono-layer step	Fluence $6.6 \cdot 10^{18}$ cm^{-2}
23.8	20.5	22.2	19.6	19

Figure Captions

Figure 1: Two-dimensional distribution of the sputtered silicon under the irradiation with a 30 keV gallium ion beam tilted 60º from the target normal measured using the TOF-SIMS method (a) and simulated using the MCSIM program (b)

Figure 2: Distribution of the sputtered silicon on a planar collector measured using the RBS for 70º (a) and 80º (b) of the 30 keV gallium ion beam tilt angle: points are for the measurement, line is for MCSIM simulation , statistical uncertainty of the measurements near the maximum is indicated by 2σ

Figure 3 Laterally integrated angular distributions of the sputtered silicon atoms simulated using different time steps DT in the XMD program for a target from isolated silicon atoms for gallium ion beam with an energy of 30 keV and a tilt angle of 80º, statistical uncertainty of the simulation near the maximum is indicated by 2σ

Figure 4 The same as in Figure 3 but for amorphous silicon sample having different atomic structure of the surface layer

Figure 5 Two-dimensional angular distribution of the silicon sputtered by a 30 keV gallium ion beam tilted by 70º, simulated using MCSIM and approximated by the analytical model suggested in this work

Figures

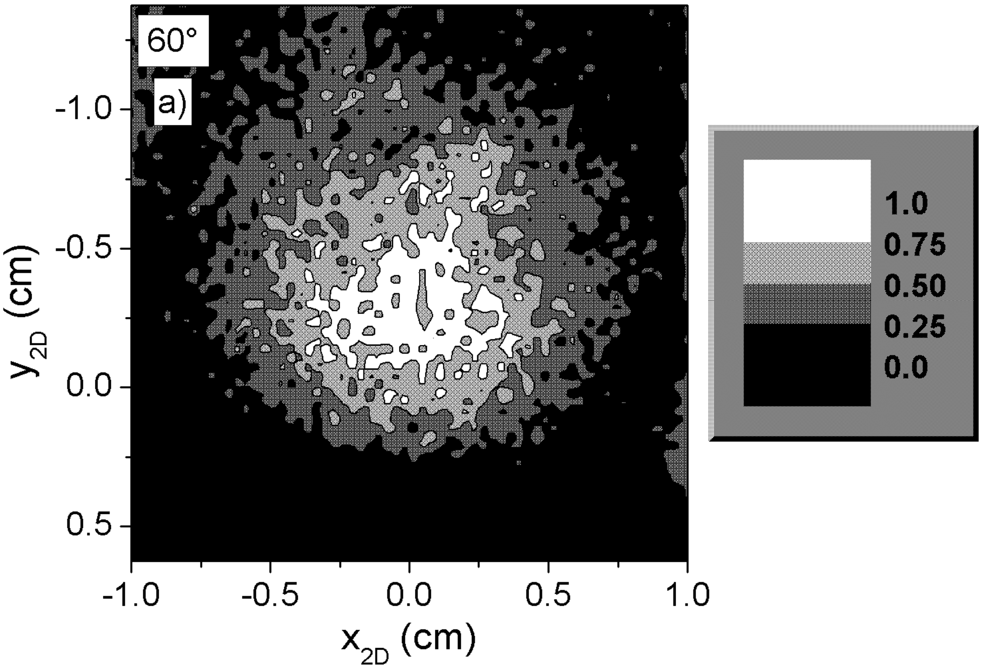

Figure 1a

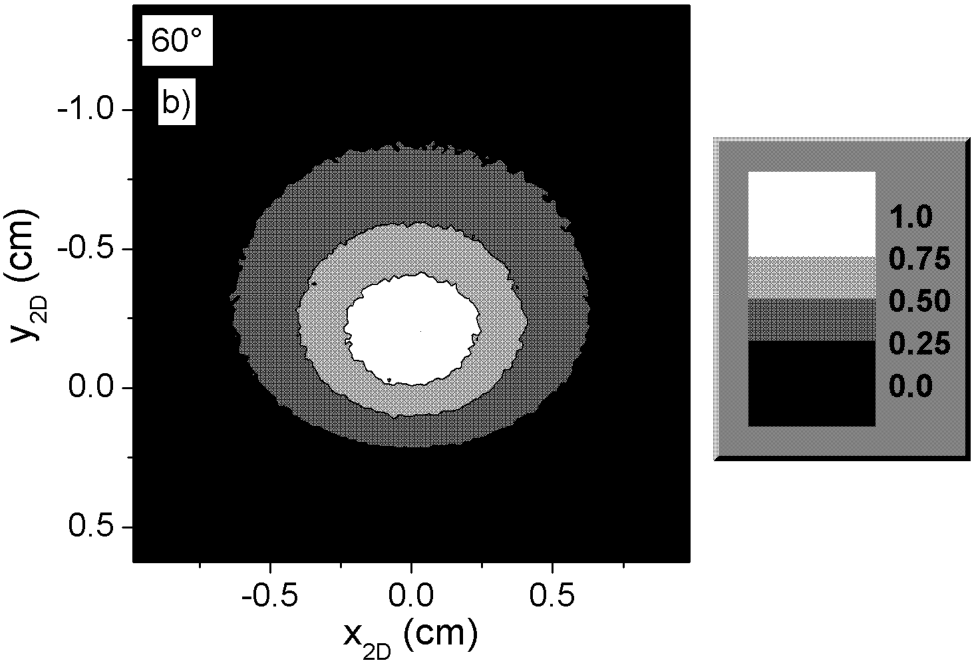

Figure 1b

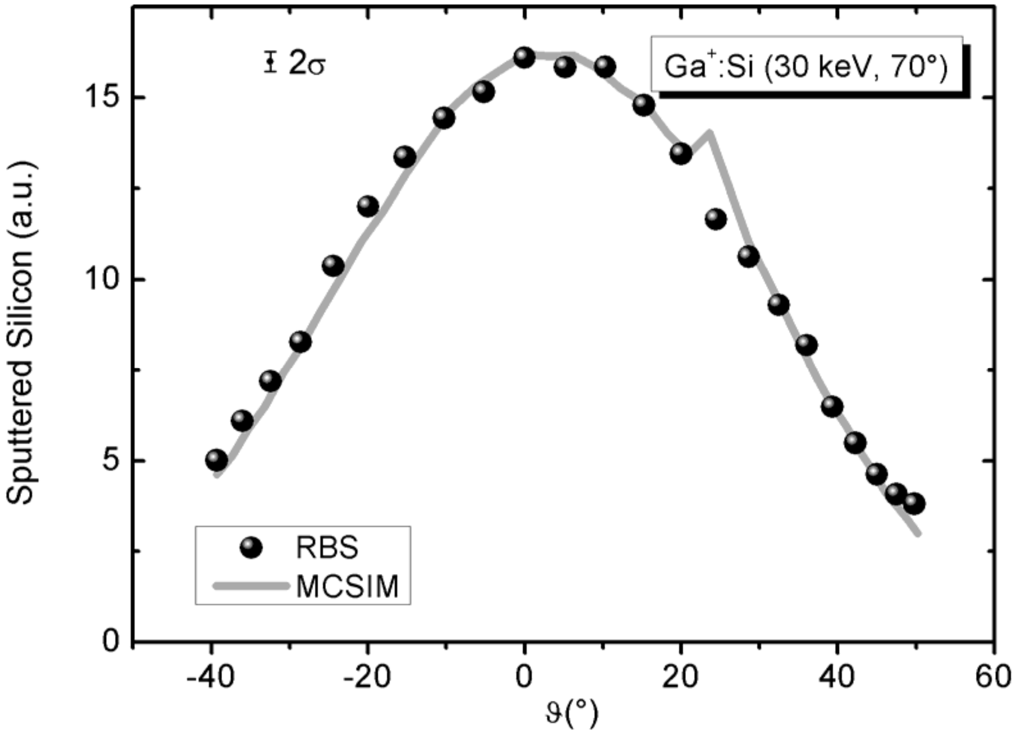

Figure 2a

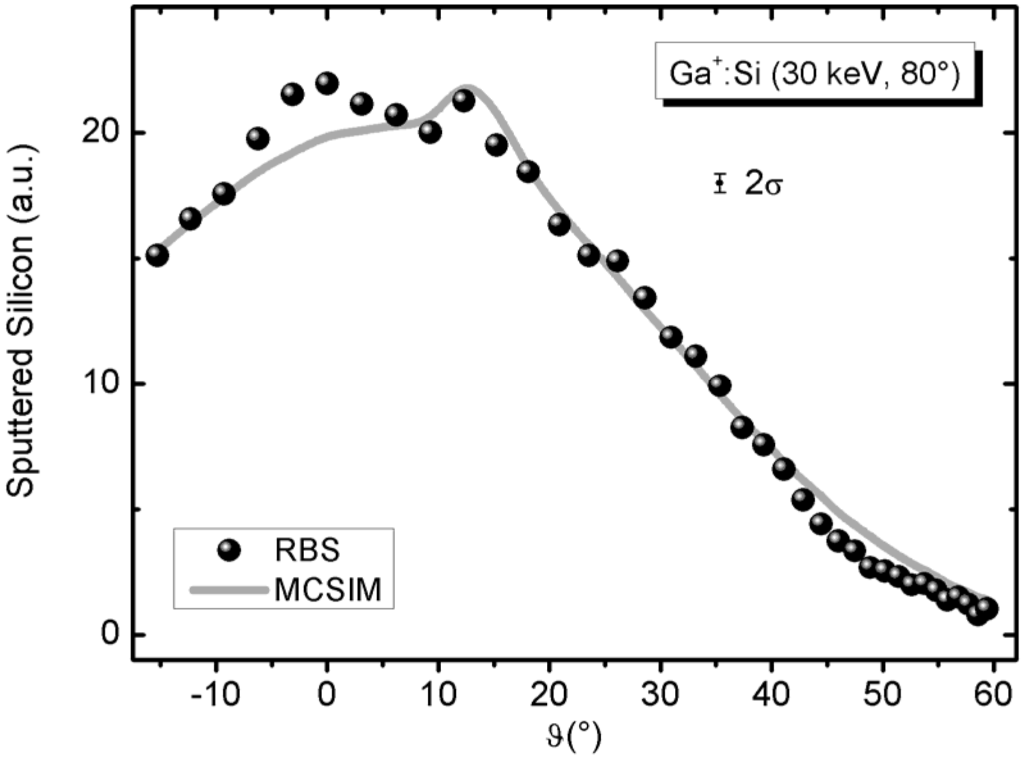

Figure 2b

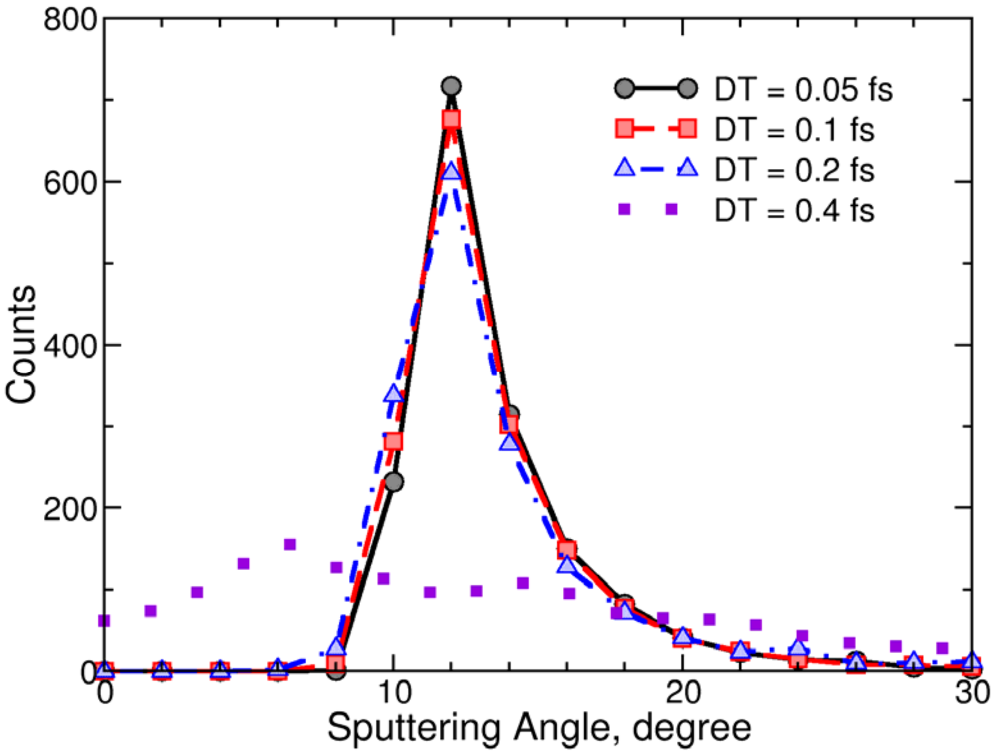

Figure 3

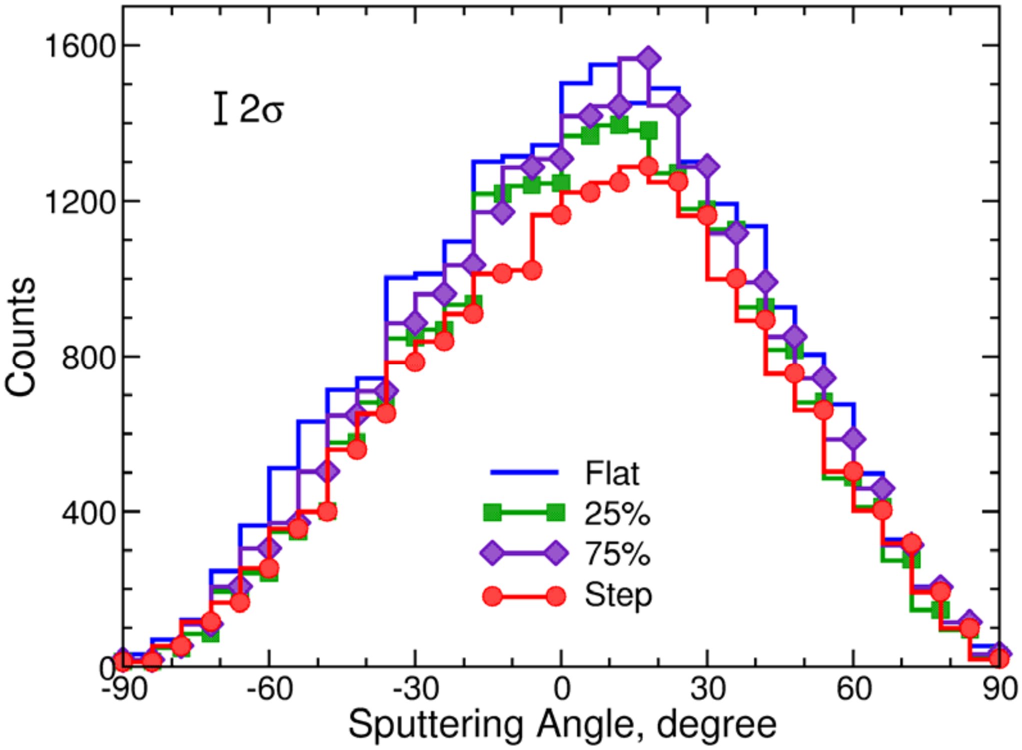

Figure 4

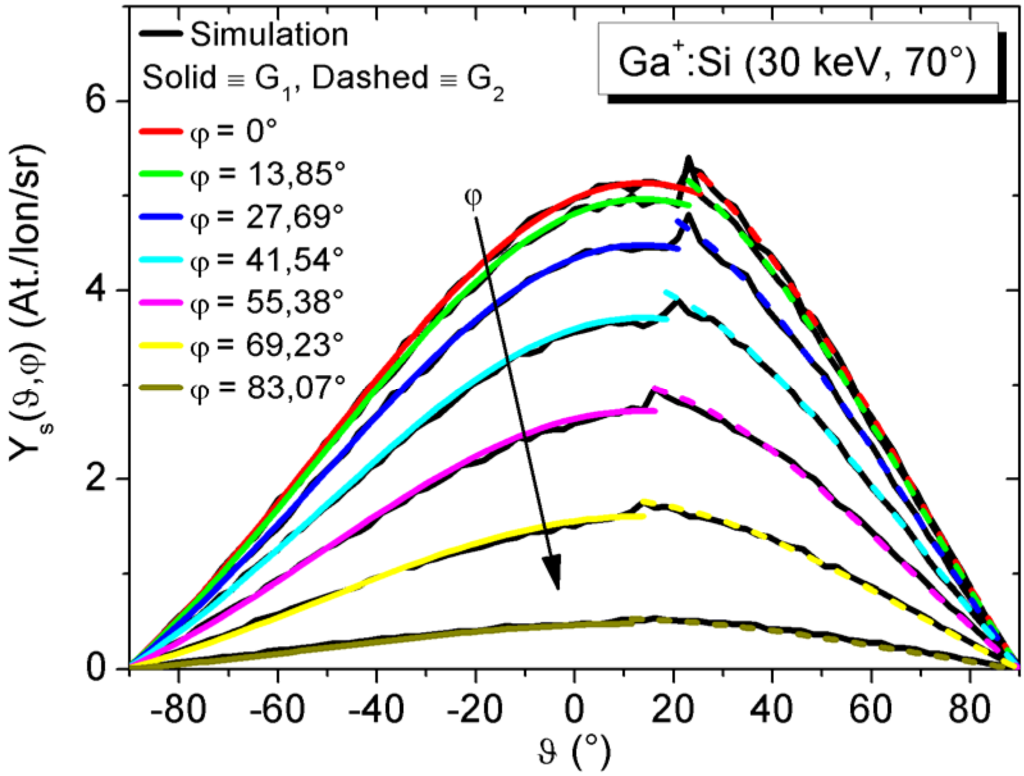
Simulation
Solid ≡ G1, Dashed ≡ G2
φ = 0°
φ = 13,85°
φ = 27,69°
φ = 41,54°
φ = 55,38°
φ = 69,23°
φ = 83,07°
Ga+:Si (30 keV, 70°)
φ
Ys(ϑ,φ) (At./Ion/sr)
ϑ (°)
0
2
4
6
-80
-60
-40
-20
0
20
40
60
80

Figure 5