Full length article

**Sputter yields of rough surfaces: Importance of the mean surface inclination angle from nano- to microscopic rough regimes**

C. Cupak, P.S. Szabo, H. Biber, R. Stadlmayr, C. Grave, M. Fellinger, J. Brötzner, R.A. Wilhelm, W. Möller, A. Mutzke, M.V. Moro, F. Aumayr

Abstract

The roughness of a surface is known to have a strong influence on the sputtering process. Commonly used 1D Monte Carlo codes for calculating sputter yields show good agreement with experimental data only for comparably flat surfaces, whereas local ion incidence angles, shadowing and redeposition influence the sputter yields in both magnitude and angular dependence on rough surfaces. In the present work, we therefore investigated tungsten samples of largely different roughness, characterised by atomic force and confocal microscopy. A highly sensitive quartz crystal microbalance was used to determine sputter yields during ion irradiation. Low ion fluences were applied to ensure that the surface morphology did not change during irradiation. Low ion fluences were applied to ensure that the surface morphology did not change during irradiation. The results were used to benchmark our new ray-tracing simulation code SPRAY, which can take microscopy images without limitations in size as input. SPRAY was furthermore applied to perform systematic simulations for artificially roughened and computer-generated surfaces. A clear result was that the governing parameter for description of the sputtering behaviour is the mean value of the surface inclination angle distribution, rather than the commonly used root mean square roughness. Our simulations show that this parameter is universally applicable for a wide range of different surface structures.

1. Introduction

Sputtering of surfaces by ion bombardment is a current topic of high relevance in surface technology and applied sciences: Physical Vapour Deposition (PVD) methods employ sputtering to create nm thin coatings for application in semiconductor industries [1], to enhance wear resistance [2] or to change optical or electro-chemical properties [3,4]. Furthermore, Focused Ion Beam (FIB) cutting methods utilise localised sputtering to modify materials in the nanometre regime [5], while nano patterning techniques like ion lithography can be used to create laterally larger and oriented surface patterns [6]. Sputtering effects can also occur in outer space on objects which do not possess a protecting atmosphere, where impinging solar wind ions lead to continuous erosion as part of space weathering [7–10]. In nuclear fusion research, erosion of first wall materials due to sputtering is under continuous examination, since sputtering by fuel or seeding gas ions limits the lifetime of plasma facing components and also leads to increased plasma impurity abundances, which are disadvantageous for the performance of fusion reactions [11–15].

Well-known dependencies of the sputter yield (defined as the mean number of sputtered atoms per incoming ion) on parameters like ion energy, ion incidence angle or the atomic mass of the target atoms can be satisfactorily described by theoretical concepts which have been established for decades [16,17]. Furthermore, established Binary Collision Approximation (BCA) simulation codes like SRIM [18], TRI-DYN [19] or SDTrimSP [20], which simulate ion-induced collision cascades, are available for sputter yield prediction. However, significant deviations between simulated and experimental results can be found under certain conditions, which often originate from complex surface properties demanding more detailed modelling. For instance, Schlueter et al. recently showed that the sputter yield of polycrystalline targets can substantially deviate from numerical results based on amorphous target models, which are often employed in simulation codes [21]. Furthermore, it is also well known that properties like surface roughness can severely change the effective sputter yield in comparison to flat surfaces [22–28].
On the one hand, this motivated alternative implementation of roughness in 1D simulation codes, for example by using fractal geometry [29] or density gradient models [30]. On the other hand, complex surface patterns may not always be satisfactorily described by such models. State-of-the-art 3D BCA codes like SDTrimSP-3D [31] or TRI3DYN [32] are advantageous as they provide the opportunity for direct surface topography implementation from microscopy, which is especially useful for simulation of complex structures like, e.g., tungsten-fuzz [33]. Modern codes like ERO2.0 furthermore enable consideration of magnetic forces in the plasma of fusion devices for particle transport [34]. Recent studies which employed such 3D simulation codes support the hypothesis, that enhanced roughness often leads to lower effective sputter yields and also affects the angular emission distribution of sputtered atoms and reflected ions [33-36]. However, many 3D codes typically demand computational cluster infrastructure and/or are limited to surface areas of comparably small lateral image size (several 100 nm) which prevents investigation of larger scale roughness effects. In addition, dynamic surface modification cannot be neglected in most experimental studies, making it difficult to assess static sputtering properties of a rough surface. Common parameters like e.g., Root Mean Square (RMS) roughness, which is defined as the quadratic mean value of vertical deviations with respect to the average surface height, are often used for characterisation of real surfaces, while it is not clear whether this parameter is suitable for a consistent assessment of sputtering.

The works of Küstner et al. [22], Hu et al. [37], Stadlmayr et al. [38] and Arredondo et al. [26] highlighted that the Surface Inclination Angle Distribution (SIAD) (see supplementary material Section A) is of high relevance for understanding rough surface sputtering by ion irradiation, since it is connected to effects like variation of local ion incidence angles, geometrical shadowing, redeposition or secondary sputtering due to reflected ions (see Fig. 1). In this work we demonstrate that the first moment (or mean value) $\delta_\mu$ of this distribution is already a very good and scale-independent parameter to characterise non-dynamic sputtering of rough surfaces. The applicability for a wide range of different surfaces was investigated experimentally using the highly sensitive QCM technique established at TU Wien. With 2 keV $\text{Ar}^+$ ions from a mass-filtered low-flux ion source, quasi-static sputter yields of W samples with roughness values ranging from the nano- towards the micrometre RMS regime were investigated (Section 3). In addition, these results were used to benchmark our newly developed ray-tracing simulation code SPRAY, which enables rapid calculation of effective sputter yields considering complete microscopy images without lateral size limits or demands for computational cluster infrastructure. SPRAY was then used for artificially roughened and computer-generated sample surfaces to investigate the role of different surface roughness parameters (Section 4).

2. Materials and methods

2.1. Tungsten samples

In the course of this study, three individual samples were used and characterised prior to the QCM experiments. The samples labelled $S_1$ and $S_2$ consisted of tungsten-coated quartz crystals with a diameter of 14 mm. The virgin quartz crystals were manufactured by the company KVG Quartz Crystal Technology GmbH, Germany. The top layer of tungsten with a thickness of several 100 nm was deposited via PVD in a collaboration with Max Planck Institute of Plasma Physics, Garching, Germany, using magnetron sputtering. While sample $S_1$ showed a very flat and mirror-like surface, $S_2$ intentionally exhibited higher roughness and a matte optical appearance. This was achieved by using quartz crystal substrates with different surface finishes, being either polished or not. Therefore, the roughness of the deposited tungsten layers was strongly affected by the topography of the underlying interface. The third sample labelled $S_3$ was produced via Chemical Vapour Deposition (CVD) in a collaboration with Forschungszentrum Jülich, Institute of Energy and Climate Research, Germany. A tungsten layer with about 700 μm thickness was deposited on a 1 mm thick silicon wafer platelet with $10 \times 10 \text{mm}^2$ size. Using this approach, a very high surface roughness was achieved. The silicon wafer was used as a substrate since the elevated temperatures of 870 K during the CVD process would have triggered a non-reversible phase transition in a quartz crystal [39]. Since surface roughness is a key property in this study, microscopy investigations were performed before and after the experiments. Tapping-mode Atomic Force Microscopy (AFM) is a versatile technique for imaging nanoscale surface features and was therefore of interest for the smoother samples in this campaign [40]. Our device Cypher S acquired from Asylum Research, Oxford Instruments (United Kingdom) enables to record images with up to 1024 $\times$ 1024 $\mu$m$^2$ resolution and 20 $\times$ 20 $\mu$m$^2$ lateral size. However, in order to enable mapping of nanoscale features a maximum image size of 2 $\times$ 2 $\mu$m$^2$ was utilised in this study. In addition, also Con-Focal Microscopy (CFM) was employed especially for investigation of the rougher samples, which could not be investigated by AFM. The $\mu$surf explorer microscope manufactured by NanoFocus AG, Germany was used to generate images with 512 $\times$ 512 $\mu$m$^2$ [41]. Using a 100x objective, a lateral image size of 160 $\times$ 160 $\mu$m$^2$ was achieved. Based on quantitative information from the obtained microscopy images, both the RMS roughness value and $\delta_\mu$ were calculated for characterisation.

The results of roughness investigation are collected in Fig. 2. The three columns from left to right indicate data from sample $S_1$, $S_2$ and $S_3$, respectively. In the top row of Fig. 2, selected AFM microscopy images of the samples are visualised in 3D using the software Meshmixer (Autodesk Inc.) [42]. The second row includes analogue visualisations like in the top row, but which are based on CFM. The third row shows diagrams of the SIAD obtained by all recorded images for a specific sample. Only AFM was applied for sample $S_1$, since the nanoscale topography could not be resolved with CFM. A flat surface with round scales of nm-height can be seen in the 1 $\times$ 1 $\mu$m$^2$ image (Fig. 2aa). In total, four images from different locations on this sample were recorded and used for calculation of roughness parameters. The nanoscale smooth surface is indicated by a small RMS roughness value of 1.7 ± 0.1 nm and is also characterised by the shape of the SIAD, which shows a pronounced peak for angles close to surface normal direction (Fig. 2ca). A mean value $\delta_\mu$ of 7.6 ± 1.2° was determined from this distribution.

Sample $S_2$ was investigated with AFM and CFM, therefore selected images are shown for both methods (Fig. 2ab and 2bb). The 2 $\times$ 2 $\mu$m$^2$ AFM image shows a corrugated surface with pronounced inclinations and superposition with round scales. Since the lateral size of the AFM...
Fig. 2. Collection of results regarding sample characterisation. The surface visualisation was created using the software Meshmixer [42]. Columns from left to right: Sample S1–S2–S3. Top row: 3D visualisation of sample surface topographies based on AFM, if available. Second row: 3D surface visualisation based on CFM, if available. Third row: Results regarding the surface inclination angle distribution (SIAD).

images was already comparable to the height of surface features, it was assumed that their representativeness is limited. Therefore, a larger set of 15 AFM images was used to gain more data for the determination of roughness parameters, which were 34.5 ± 26.9 nm for RMS and 19.7 ±4.2° for $\delta_{m}$. The comparably large standard deviation in RMS corroborates the assumption, that the recorded AFM images were not a reliable basis for RMS calculation. Furthermore, any superimposed surface structures which exist on a larger lateral scale, would have been hard to include with these images. Nevertheless, the AFM images of sample S2 revealed important characteristics of the nanoscale topography on the locally investigated surface sections. In contrast to the results of AFM, the same sample surface exhibits smooth scales accompanied by some circular pores in the corresponding 160 × 160 $\mu$m² CFM image. A better representativeness of CFM images may be assumed due to their significantly larger lateral size, but the resolution was not high enough to map the local nanoscale features found in the AFM images. Resulting differences can also be seen in the SIAD tendencies, where CFM based results (blue line) suggest characteristics of a smooth surface similar to the data of S1, while the AFM based results (red line) show more contributions from tilted surface features (Fig. 2cb). The values of roughness parameters based on five CFM images were determined with 187 ± 5 nm for RMS and 11.2 ±0.2° for $\delta_{m}$. It is important to mention that the smaller value of $\delta_{m}$ was mainly attributed to the lack of CFM resolution, which resulted in artificial smoothing of the topography. Similar limitations were observed by Kelemen et al. who utilised confocal microscopy to generate input for SDTrimSP-3D [28]. In contrast, the larger lateral image size of CFM allowed to detect surface features on a larger scale, resulting in a higher RMS value.

Sample S3 was only investigated using CFM, since it was too rough for AFM. A corrugated topography with surface features of several $\mu$m height was found (Fig. 2bc). Five CFM images were used for roughness parameter determination, resulting in 1551 ± 48 nm for RMS and 36.5 ±1.2° for $\delta_{m}$. The corresponding SIAD shows a broad width, indicating that locally substantial surface tilts are present on this sample (compare Fig. 2cc).

Considering the wide range of RMS and $\delta_{m}$ values between the samples, our set of experimental specimens provided a basis to investigate roughness-induced sputtering effects from the nano- towards the microscale roughness regime. Consideration of additional roughness parameters like for example the autocorrelation length or the hurst parameter were beyond the scope of this study [43]. The recorded microscopy images were furthermore used for numerical simulations using the code SPRAY (see Section 2.3). The complete set of images
can be inspected in the supplementary material Section E. All samples were also investigated by AFM or CFM after the ion beam experiments, but no qualitative difference in the topography was observed. Since the determined roughness parameters were furthermore well within the scatter of a-priori measurements, it can be assumed that no significant surface modification occurred during the experiments. The obtained roughness parameters for all surfaces are summarised in Table 1 in Section 3 for comparison with measured and simulated sputter yields.

In order to validate the purity of the tungsten layers, depth-resolved information of the samples' composition was obtained by means of Ion Beam Analysis (IBA) [44]. Coincidence Time-of-Flight/Energy Elastic Recoil Detection Analysis (ToF-E ERDA) was carried out with 36 MeV $^1$H$^+$ as probing beam [45]. Except near surface contaminations with light impurity species like hydrogen, carbon or oxygen, a high tungsten purity of at least 98 at.\% was validated for all samples. Compositional depth profiles and more detailed information regarding IBA can be found in the supplementary material Section B. Since all QCM experiments included an in-situ Ar sputter cleaning procedure to remove surface contaminations and adsorbates, the assumption of pure tungsten surfaces can be justified in this study.

2.2. Quartz Crystal Microbalance setup

The Quartz Crystal Microbalance (QCM) is a versatile technique which allows to determine net mass variations of a quartz crystal with high precision by recording the quartz’s resonance frequency. Sauerbrey established a basis for the QCM in 1959, describing the relation between relative mass and frequency changes in form of Eq. (1) [46].

$$\frac{\Delta m}{m} = -\frac{\Delta f}{f} \tag{1}$$

Eq. (1) shows that a mass loss (or gain) results in a proportional frequency increase (or decrease). It remains valid also for quartz crystals with a thin layer of different material on top of its surface. At TU Wien, application of this technique for ion sputtering investigation has a long history and was continuously improved over the years [47–49]. For QCM samples under ion bombardment, the resulting change of the resonance frequency with time $\Delta f / \Delta t$ can be recorded via dedicated electronics. Using this frequency change, a calculation of the sputter yield $Y$ by consideration of Eq. (1), ion current density $j$ and a constant factor $C_j$ is possible. The connection is given by Eq. (2) [47].

$$Y \ [\text{atoms/ion}] = C_j \cdot \frac{1}{j} \cdot \frac{\Delta f}{\Delta t} \tag{2}$$

$$C_j = \frac{e_0 \cdot q \cdot l_Q \cdot \rho_Q}{m_n \cdot \frac{\omega}{\rho}, \Delta f / \Delta t}$$

$C_j$ can be calculated by the electron charge $e_0$, the atomic mass unit $m_n$, and the ion charge state $q$. Also the atomic mass of tungsten $m_n$ and quartz crystal parameters like density $\rho_Q$, thickness $l_Q$ and initial resonance frequency $f$ are contributing to this constant. The ion current density $j$ is determined by Faraday cup measurements prior and after each QCM irradiation. It is important to mention that pure tungsten samples are assumed in this calculation and direct sputter yield determination is only possible for tungsten layers deposited on QCM crystals like for samples S1 and S2.

For investigation of sample S3, which was not a QCM crystal but consisted of a tungsten film on a silicon wafer, we used a technique based on the work of Berger et al. [50]. Here, a QCM is used as a catcher for the sputtered material, which results in a mass gain signal during target irradiation. In our setup, this C-QCM, which also incorporates a W-coated QCM quartz, can be positioned along a circular path around the target sample in order to probe the angular distribution of sputtered particles. A scheme describing the geometrical conditions in our experimental setup is presented in Fig. 3. More detailed information regarding the utilised C-QCM technique can be found in Ref. [50] and the supplementary material Section C.

Our experimental setup consists of an Ultra High Vacuum (UHV) chamber with base pressures on the range of $10^{-9}$ mbar. The SPECS IQE 12/38 sputter ion source, equipped with a Wien-filter for mass separation, was used for creation of a low intensity 2 keV Ar$^+$ ion beam with a flux on the range of $\Phi \approx 10^{16}$ ions/m$^2$/s. A total Ar$^+$ fluence per sample on the order of $1.5 \times 10^{20}$ ions/m$^2$ was chosen to ensure that no significant surface modifications were induced during irradiation. Assuming a typical sputter yield of 2 W/Ar, this resulted in erosion of about 5 nm per sample.

A target holder was used to mount QCM crystals or other sample types centrally in the vacuum chamber (see Fig. 3). It also incorporated the Faraday cup for determination of ion beam currents. Using a 4-axes manipulator, accurate positioning and exposition of the target sample holder towards the ion beam was achieved. Most of the experiments were performed at a fixed ion incidence angle of $\Phi = 60^\circ$. The target holder allowed usage of either a QCM crystal or other targets. The C-QCM system (B) was positioned independently along a circular path around the target, maintaining a constant radial distance $r = 21.4$ mm, which enabled to probe the angular distribution of sputtered atoms and reflected ions from the target.

2.3. SPRAY simulations

Our newly developed simulation code SPRAY (SPuttering simulation via RAY tracing of particles) was used in this study to model sputtering and redeposition in the case of rough surfaces. The main advantage of this code is its ability to use microscopy images limitless of...
size for sputter yield calculation, utilising an approach similar to ray-tracing techniques in 3D rendering. The code is written in PYTHON 3.8 language, supported by the open-source package VTK 9.0 [52,53] for implementation of ray-tracing features and can be executed on conventional desktop PCs. Since this code was initially developed in the course of this study, a brief overview on its features and implemented physics is given in this section. A more detailed technical description of the code can be found in the supplementary material Section D.

The latest version of SPRAY supports roughness dependent sputter yield calculation under consideration of ion incidence angle variation, redeposition, shadowing effects and secondary sputtering from locally reflected ions. In addition, three-dimensional emission distributions of sputtered particles and reflected ions can be calculated for a rough surface. For this work only a pure tungsten surface was considered, while it is principally also possible to simulate multicomponent samples. Furthermore, simulations are limited to static calculations only, which implies that the surface topography and composition are fixed and will not vary with fluence as e.g., in the dynamic versions of SDTrimSP-3D [31] or TRI3DYN [32].

Data regarding the sputter yields of a flat surface and the corresponding angular distributions have to be provided as inputs for SPRAY, in example from BCA codes. Principally, the data origin is not limited to a certain BCA code, but can be taken liberally from any source. In this study, W sputter yields were generated by SDTrimSP [20]. In addition, three-dimensional emission distributions for sputtered atoms and reflected ions were generated with TRI3DYN [32]. This highlights a difference to other simulation codes which follow a similar strategy, where the angular distribution of sputtered or reflected particles is mostly approximated by using a cosine distribution [22,54,55].

As a next step, one or more microscopy images of the sample surface were used as input for SPRAY. In this study, the same images also provided the basis for calculation of roughness parameters RMS and $\delta_s$ (see Section 2.1). It should be mentioned that the use of SPRAY is limited to surface features exceeding the extension of the collision cascade in size. E.g., fibrous structures with a fibre thickness smaller than the mean ion range might not be properly calculated with SPRAY.

The general routine during SPRAY execution can be described by four main tasks: Primary ion impact on randomly selected surface points, simulation of local sputtering, computation of local ion reflection and consideration of secondary sputtering by reflected ions (compare Fig. 4a-d). At first, a primary ion is directed on a randomly selected impact point on the surface, while the raytracing algorithm intrinsically considers shadowing effects (Fig. 4a). For a given impact point the local incidence angle is calculated, which determines the local sputter yield from the input data. Secondly, trajectories for sputtered atoms are launched, while the raytracing algorithm determines the number of redeposited atoms (Fig. 4b). This allows to compute a redeposition factor, which scales down the local sputter yield. Originating from the same primary impact point, trajectories of reflected ions weighted by their corresponding reflection coefficient are launched using a similar approach (Fig. 4c). Special treatment of secondary ion impact points is implemented, since they provide a starting point for secondary sputtered atoms (Fig. 4d). In analogy to the simulation of primary sputtered atoms, secondary sputtering is calculated based on the local incidence angle and redeposition.

The whole routine is repeated for all simulated primary ions, which finally allows to calculate a mean effective sputter yield for a given surface and output lists with effectively sputtered particle trajectories.

3. Experimental results and comparison to simulations

For initial benchmarking, sputter yields of the samples S1 and S2 as a function of ion incidence angle $\varphi$ were determined both by QCM experiments and SPRAY simulations. A summary of the results is shown in Fig. 5. For the smooth sample S1, two measurements under ion incidence angles $\varphi = 0^\circ$ and $\varphi = 60^\circ$ were performed (green data points). Very good agreement in comparison to SPRAY simulations was found (dashed green line), where the full set of available AFM images was used. For the medium rough sample S2, the ion incidence angle $\varphi$ was varied in an interval between 0° to 70° in steps of 5°, exploiting the maximum angular range available in our experimental setup. Again, a good overall agreement between measured (black data points) and SPRAY simulated sputter yields (dashed black line) was found over the whole angular interval, with small deviations (max. 10%) at around $\varphi = 40^\circ$. In addition, a perfectly flat surface was also used as input for SPRAY. As expected, the code reproduced repository results which were obtained from SDTrimSP [20] (dashed orange line). In comparison to the results of sample S1, it can be seen that even nanometre-smooth topographies affect the sputter yield, especially for grazing ion incidence angles.

In the following, experimental C-QCM signals are presented by the data points in Fig. 6 for all samples, allowing to discuss the angular distribution of sputtered atoms. The ion incidence angle $\varphi$ was kept at 60° in this case. In addition, SPRAY simulated angular distributions of sputtered atoms are presented in form of colour-shaded areas. For this, the distributions were discretised using bins of $2.15 \times 10^{-3}$ sr size along the plane of ion incidence, before re-normalisation was applied with a constant factor (for all samples). This factor was chosen for best agreement of the angular distribution to the C-QCM signal of S1 at position $\varphi = 0^\circ$.

During ion irradiation of the smoothest sample S1, the highest experimental C-QCM signal (green) of all samples was measured within the accessible angular interval (Fig. 6a). A maximum was found in the forward-sputtering regime at $\varphi = 30^\circ$, which probably originates from single knock-on sputtering events [56]. Simulated results on the basis of AFM images (magenta shaded area) show good agreement to the experimental data. However, the forward sputtering peak appears shifted at 50°, which corresponds well to simulated results for a perfectly flat surface using TRI3DYN (compare figure 3 in the supplementary material Section D). For the rougher samples, the measured signals were generally lower and the single knock-on peak was not found.

Fig. 6b shows the comparison for sample S2, with SPRAY simulation results on the basis of both AFM (magenta) and CFM images (orange). Especially the AFM based data set is located well within the error bars of the experimental data (black), while a clear offset between the simulated data sets is visible. The CFM data generally shows a higher trend than the AFM based data and indicates the importance of highly resolved images for SPRAY simulations. In fact, the tendency of the CFM based data is comparable to the results for the smoother sample S1, in accordance to the similar appearance of microscopy images (see Fig. 2).

The results for the roughest sample S3 show a measured C-QCM signal (red) with a very collimated orientation along the sample surface normal direction at 0° (see Fig. 6c). A similar result was also found by Eksaeva et al. [34] and can be explained by a reduced number of successfully sputtered atoms escaping in a tilted direction respective to the surface normal due to a higher probability for local redeposition. Lower C-QCM signals indicate that the sputter yield decreased for this rougher surface. The simulated angular distribution shows good agreement to the experimental data with respect to the orientation, while the experimental values between $-10^\circ$ to $0^\circ$ appeared to be slightly higher.

Since the samples S1 and S2 were QCM quartzes, also direct determination of sputter yields was feasible during the experiments. For S1, the highest sputter yield of $2.30 \pm 0.23$ W/Ar of all samples was found, while a decreased sputter yield of $1.95 \pm 0.19$ W/Ar was measured for the medium rough sample S2. Since for sample S3 no direct evaluation of the sputter yield was possible, an estimation procedure was applied by using the results for sample S2 as a reference. Polynomial functions of maximum order 3 were fitted to the C-QCM data sets in Fig. 6. Then, the integral of each fit function within the accessible angular interval was calculated. Finally, the ratio of integrals
Fig. 4. Simulation scheme for SPRAy. (a) Primary ion (red) hits the surface on a randomly selected impact point (yellow asterisk), enabling choice of local sputter yield from the input data via the local incidence angle $\phi$. (b) Raytracing of trajectories for sputtered atoms (black) is initiated. Sputtered target atoms which hit the surface again are considered as redeposited. (c) Raytracing of trajectories for reflected ions (red) is initiated. (d) Secondary sputtering by reflected ions is calculated, by consideration of specific input data set for the local ion impact. The whole procedure is repeated for every simulated primary ion.

Fig. 5. Comparison of sputter yield values $Y$ from QCM experiments (points) and SPRAy simulations (dashed lines) under variable ion incidence angle $\phi$ for samples S1 (green) and S2 (black). The SPRAy simulation for a flat surface (orange dashed line) reproduces the repository data results from SDTrimSP.

between the samples S3 and S2 was multiplied with the reference sputter yield. Via this approach, a value of $1.51 \pm 0.40$ W/Ar was calculated for S3, while this procedure introduced a comparably large uncertainty. For completeness, the sputter yield of S1 was also deduced this way ($2.38 \pm 0.03$ W/Ar), which agrees well with the directly measured value. All experimental sputter yields are listed in Table 1 for comparison.

The SPRAy simulations also provide sputter yield results based on all available microscopy images, which allowed to assess standard deviations to the numerical values. For samples S1 and S2, simulated sputter yields for the AFM based cases ($2.29 \pm 0.03$ W/Ar and $1.95 \pm 0.14$ W/Ar, respectively) indicated excellent agreement with experimental data, which was already shown in Fig. 5. However, the simulated value of $2.30 \pm 0.01$ W/Ar based on CFM images of sample S2 is comparable to the results of sample S1, which corresponds well to the similarity in their roughness parameter $\delta_{\text{rms}}$ deduced from the utilised microscopy images. It has to be noted, that at the same time the RMS roughness values of these two samples differ by a factor of 100. For sample S3, SPRAy simulations on the basis of CFM images resulted in a sputter yield of $1.53 \pm 0.04$ W/Ar, in good agreement to the experimentally estimated value. This highlights, that for microscopically rough samples an application of CFM can still be a viable option.

Considering the trends in sputter yields and roughness parameters in Table 1, a general tendency can be seen that higher surface roughness may lead to a decreased sputter yield. Still, the case of sample S2 indicates that RMS roughness is not a suitable parameter for sputter yield estimation and that the utilised microscopy method strongly influences the results of SPRAy and roughness parameter determination. The simulated results regarding sample S1 and S2 further indicate that similar $\delta_{\text{rms}}$ values can be connected to similar sputter yields and provide a better basis for empirical modelling. However, the low number of available experimental samples and data does not yet allow to draw fundamental conclusions regarding the relation between the sputter yield and $\delta_{\text{rms}}$ or the universality of this approach. Still, the results of this benchmark study show that SPRAy is a versatile tool for detailed investigation of roughness related sputtering effects and that good prediction of sputter yield values can be obtained if suitable microscopy images are taken as input. In order to further investigate the applicability of roughness parameter $\delta_{\text{rms}}$, the following Section 4 is focusing on a dedicated SPRAy investigation which allows to corroborate the results from this benchmark.

4. Systematic simulations with artificially generated rough surfaces

In order to further investigate the role of roughness parameters RMS and $\delta_{\text{rms}}$ for sputtering, a more comprehensive numerical study with SPRAy was performed. For this purpose, not only microscopy images of real samples, but also artificial computer-generated surfaces were used as input. At first, the original CFM images of samples S2 and S3
\[ \mu \text{ whole RMS range between 0 and 1} \]

were evaluated regarding RMS and than 1.0 corresponds to enhanced roughening. All surfaces of such a set than 1.0 therefore led to artificial smoothing, while a factor bigger creating roughness. In the following, the original surfaces' height values were multiplied with a scaling factor. Application of a factor smaller of a constant factor for all three samples.

re-normalisation of these simulated angular distributions was performed by application hereinafter called \( S2-s \) already very rough on the microscale. These two surface data sets are \[ \delta \text{ RMS AFM [nm]} - 1.7 \pm 0.1 \quad 34.5 \pm 26.9 \quad - \]
\[ \delta \text{ RMS CFM [nm]} - 187 \pm 5 \quad 1551 \pm 48 \]
\[ \delta \text{ CFM [deg]} - 11.2 \pm 0.2 \quad 36.5 \pm 0.6 \]
\[ \text{exp. QCM Y [W/Ar]} - 2.30 \pm 0.23 \quad 1.95 \pm 0.19 \quad - \]
\[ \text{exp. est. C-QCM Y [W/Ar]} - 2.38 \pm 0.59 \quad 1.95 \pm 0.19 \quad 1.51 \pm 0.40 \]
\[ \text{SPRAY Y AFM [W/Ar]} - 2.29 \pm 0.03 \quad 1.95 \pm 0.14 \quad - \]
\[ \text{SPRAY Y CFM [W/Ar]} - 2.30 \pm 0.01 \quad 1.53 \pm 0.04 \]

In addition, a set of artificial topographies generated by a computer algorithm was introduced, which are labelled G in the following. Their individual height values were chosen from a Gaussian distribution on each point of an equally spaced lateral grid with \( 5 \times 5 \mu m^2 \) side length and 50 nm spacing, following the same approach as Li et al. [35]. The desired RMS values were generated in a range between 0.0 and 0.5 \( \mu m \). The roughest surface generated this way already showed very pronounced needle-like features, appearing similar to the beryllium surface structures found after deuterium plasma exposure in a study by Doerner et al. [57] or to the computer-generated structures used by von Toussaint et al. [31]. A more detailed description of the surface preparation and an overview over all utilised topography sets can be found in the supplementary material Section F.

As a starting point, all surfaces were used in SPRAY simulations for investigation of sputtering due to 2 keV Ar\(^+\), for two selected cases of fixed ion incidence angle \( \varphi = 0^\circ \) (normal incidence) and \( \varphi = 60^\circ \). The former is of special interest, since for this case, shadowing effects can be neglected for our set of investigated surfaces. In contrast, the latter case corresponds to the same situation as in our C-QCM experiments presented in Section 3.

The results for the \( \varphi = 0^\circ \) case are presented via scatter plots in Fig. 7(a) and (b), which show the simulated sputter yields versus RMS or \( \delta_m \), respectively. A perfectly flat surface is indicated by a value of 0.0 for both RMS and \( \delta_m \). As can be seen in Fig. 7(a), RMS is not a good roughness parameter, since individual sputter yield tendencies deviate significantly for different surface sets. Especially for surfaces used in the data set G, corrugated structures with substantial inclinations were generated already for selection of relatively low RMS values, which resulted in redeposition effects on the sputter yield. However, this cannot be observed for surfaces of the other data sets with similar RMS value. Characterisation with \( \delta_m \) shows better agreement (see Fig. 7b), especially for small values below \( \delta_m < 20^\circ \). In this region, a moderate increase of the sputter yield was found, starting to rise from a value of 1.38 W/Ar for a flat surface to 1.5 W/Ar (or about +10\%) at \( \delta_m = 20^\circ \). This can be explained by a corresponding increase of the effective ion incidence angle, which has a similar effect as for a flat surface being irradiated under an ion incidence angle of \( \varphi = \delta_m \) (orange dashed line). This allows for a rough approximation in this regime without need of simulations. Beyond \( \delta_m = 20^\circ \), the data becomes more scattered, while a clear reduction of the sputter yield becomes visible for larger values of \( \delta_m \) due to redeposition of sputtered atoms. Here, the approximation via the flat surface sputter yield \( Y(\varphi = \delta_m) \) is no longer possible.

In Fig. 7(c) and (d), the results for an ion incidence angle \( \varphi = 60^\circ \) are shown. Again, RMS did not allow a satisfying characterisation of sputtering (see Fig. 7c). In contrast, quite a universal behaviour for all simulated surfaces is found when using \( \delta_s \) as roughness parameter (see Fig. 7d). For very small values of \( \delta_s < 4^\circ \), the sputter yield values remained on a level of 2.45 W/Ar, before a continuous decrease is found. The data indicates that between \( \delta_s = 4^\circ \) and \( \delta_s = 70^\circ \) a linear dependence of the sputter yield on \( \delta_s \) can be assumed, which

---

**Table 1**

List of roughness parameters (RMS, \( \delta_m \)) and sputter yields (experimental results & SPRAY simulations) for the benchmark test case. Irradiation was performed with 2 keV Ar\(^+\) on W under 60\(^\circ\) incidence angle; Usage of both AFM and CFM was only possible for sample S2.

<table>
<thead>
<tr>
<th></th>
<th>S1</th>
<th>S2</th>
<th>S3</th>
</tr>
</thead>
<tbody>
<tr>
<td>RMS AFM [nm]</td>
<td>1.7 ± 0.1</td>
<td>34.5 ± 26.9</td>
<td>-</td>
</tr>
<tr>
<td>RMS CFM [nm]</td>
<td>-</td>
<td>187 ± 5</td>
<td>1551 ± 48</td>
</tr>
<tr>
<td>( \delta_m ) AFM [deg]</td>
<td>7.6 ± 1.2</td>
<td>19.7 ± 4.2</td>
<td>-</td>
</tr>
<tr>
<td>( \delta_m ) CFM [deg]</td>
<td>-</td>
<td>11.2 ± 0.2</td>
<td>36.5 ± 0.6</td>
</tr>
<tr>
<td>exp. QCM Y [W/Ar]</td>
<td>2.30 ± 0.23</td>
<td>1.95 ± 0.19</td>
<td>-</td>
</tr>
<tr>
<td>exp. est. C-QCM Y [W/Ar]</td>
<td>2.38 ± 0.59</td>
<td>1.95 ± 0.19</td>
<td>1.51 ± 0.40</td>
</tr>
<tr>
<td>SPRAY Y AFM [W/Ar]</td>
<td>2.29 ± 0.03</td>
<td>1.95 ± 0.14</td>
<td>-</td>
</tr>
<tr>
<td>SPRAY Y CFM [W/Ar]</td>
<td>-</td>
<td>2.30 ± 0.01</td>
<td>1.53 ± 0.04</td>
</tr>
</tbody>
</table>

---

Fig. 6. Comparison between the experimental C-QCM signal (points with errorbars) and the simulated angular distribution of sputtered atoms from SPRAY (shaded areas) based on AFM (magenta) or CFM (orange) images. The angle of 0\(^\circ\) corresponds to the surface normal direction of the irradiated sample. (a) Results for the smoothest sample S1; (b) Results for the medium rough sample S2; (c) Results for the roughest sample S3. The simulated angular distributions were created by angular binning with a size of \( 2.15 \times 10^{-3} \) sr within the plane along ion incidence. For visualisation, consistent re-normalisation of these simulated angular distributions was performed by application of a constant factor for all three samples.

were used to produce sets of surface topographies with continuously increasing roughness. In the following, the original surfaces' height values were multiplied with a scaling factor. Application of a factor smaller than 1.0 therefore led to artificial smoothing, while a factor bigger than 1.0 corresponds to enhanced roughening. All surfaces of such a set were evaluated regarding RMS and \( \delta_m \) for roughness characterisation. The original surface of S2 was scaled in an interval from 0.0 to 1.0, using a step size of 0.1 for the scaling factor in order to cover the whole RMS range between 0 and 1 \( \mu m \). For the surface of S3, only an interval from 0.0 to 1.0 was used, since the original topography was already very rough on the microscale. These two surface data sets are hereinafter called S2-s and S3-s.
brown data was simulated using computer generated surfaces with Gaussian distributed height values. (a) Sputter yield $Y$ versus RMS $\delta$ characterised either by RMS or by $\phi$ for surface, allowing estimations within an error of $\pm0.05$ W/Ar, independent of $\phi$. The sputter yield $Y$ and the mean inclination angle $\delta$ of the sputtered surface under normal incidence, $\phi=0^\circ$, resulted in higher sputter yields. Similar as for the results in Fig. 7b, this can be explained by enhanced local sputtering due to favourable incident angle conditions, while redeposition effects do not yet contribute to severe reductions.

For higher $\delta_m$ values beyond $50^\circ$, the sputter yields decline continuously and settle below 1 W/Ar. An additional decrease of the sputter yield was found for irradiation with ion incidence close to the surface normal direction in combination with simulated surfaces having high $\delta_m$ values, which originated from a very spiked and corrugated topography. If an ion impinges on such a rough topography under surface normal direction, sputtering can occur on highly inclined surface facets only. Under such conditions, the emission distribution of sputtered particles has a strong forward orientation, which supports redeposition on other surface features and strongly reduces the sputter yield, similar to results found in literature [33,35].

The results in Fig. 8 substantiate the hypothesis that $\delta_m$ can be a promising parameter for description of rough surface effects on sputtering for a wide range of topographies. The computer-generated data set G, which covered the whole parameter range for both ion incidence angle $\phi$ and $\delta_m$, furthermore enabled us to develop a fit formula (see Eq. (4)) for the sputter yield of rough W surfaces under 2 keV Ar$^+$ irradiation. This fit function, which is indicated by the black dots in Fig. 8b, incorporates three parameters: ion incidence $\phi$, roughness parameter $\delta_m$, and the sputter yield of a flat W surface under normal incidence, $Y_0=1.38$ W/Ar. The latter value was again obtained from $SDTrimSP$. In total, a set of seven fit parameters...
was necessary to achieve satisfactory fitting towards the simulated data, which are listed in Table 2. Subfunction \( A(\psi, \delta_m) \) in Eq. (4) describes the sputter yield dependence on the ion incidence angle \( \psi \) for smooth surfaces with \( \delta_m = 0 \). This part is motivated by a Taylor series expansion of an empirical formula developed by Yamamura et al. [22,58]. The exponential function reduces the contribution of \( A(\psi, \delta_m) \) for higher values of \( \delta_m \). Subfunction \( B(\psi, \delta_m) \) is introduced in order to fit the data in the regime with high \( \delta_m \) and normal ion incidence, where large amount of redeposition occurs. Finally, subfunction \( C(\psi, \delta_m) \) introduces simple polynomial dependencies to achieve a good overall shape. The results show good agreement to the simulated data, except for recognisable deviations for rougher surface structures in the \( \psi \geq 80^\circ \) ion incidence regime. It can be assumed that a similar fit may also be found for other ion-target combinations, as long as kinetic sputtering remains the dominant mechanism for particle ejection.

Different results from this numeric study may be obtained if idealised surfaces like e.g., pyramidal or sawtooth profiles are used, or if anisotropic structures are investigated. Deviations are also expected for scenarios where the sputtered atoms are not emitted along straight trajectories, which was out of the scope of this study. In example, magnetic forces inside the plasma of nuclear fusion devices cause gyration of sputtered and ionised atoms. This can redirect emitted atoms towards the surface, introducing further enhancement of redeposition [12].

\[
Y(\psi, \varphi, \delta_m) = Y_0 \cdot \left[ f_1 \cdot A(\psi, \delta_m) \cdot B(\psi, \delta_m) + C(\psi, \delta_m) \right]
\]

(4)

\[
A(\psi, \delta_m) = 1 + \left( f_2 \cdot \varphi^2 + f_3 \cdot \varphi^4 \right) \cdot \exp \left( -\frac{\delta_m}{f_4} \right)
\]

\[
B(\psi, \delta_m) = 1 - \exp \left( -\frac{\varphi^2 + (90^\circ - \delta_m)^2}{f_5} \right)
\]

\[
C(\psi, \delta_m) = f_6 \cdot \varphi + f_7 \cdot (90^\circ - \delta_m)^2
\]

5. Summary and conclusion

The main goal of this study was to systematically investigate the influence of surface roughness on static sputtering and to identify which roughness parameter is suitable to characterise sputtering behaviour of corrugated surfaces. As a test case, three different W targets with surface topographies ranging from nano- towards microscopic roughness were investigated during 2 keV Ar\(^{+}\) bombardment. ToF-E ERDA was performed to validate the purity of the tungsten samples, while both AFM and CEM investigations allowed to characterise the specimens in terms of two selected roughness parameters, being the well-known RMS value and also the newly introduced \( \delta_m \) value. The latter corresponds to the mean value of the Surface Inclination Angle Distribution (SIAD) with respect to the global surface normal direction. QCM experiments allowed both to measure target sputter yields and to probe the angular distribution of sputtered atoms simultaneously and in-situ for each individual sample. A low ion flux ensured that no severe surface modifications were introduced, allowing to neglect dynamic effects. In addition, the new simulation code SPRAY was presented, which can be used to calculate effective sputter yields of rough surfaces. Key features of SPRAY are the opportunity to utilise microscopy images without limitations in lateral size and an efficient ray-tracing algorithm which is used to extrapolate \( \text{a-priori} \) simulated BCA code results of flat surfaces. Using this ray-tracing approach, effects like local ion incidence variation, shadowing, redeposition or secondary sputtering were successfully implemented. The data from QCM experiments was then used for a benchmark of SPRAY. Even though small deviations were identified regarding the angular distribution of sputtered atoms, a very good agreement was found for the effective sputter yields, well justifying its further application. Results from these experiments and simulations confirm that in the case of \( \psi = 60^\circ \), the sputter yield becomes lower for increasing surface roughness, which is in general accordance with literature. However, the experimental data from three targets were not sufficient to draw a firm conclusion regarding the question which roughness parameter is better suited to characterise sputtering of rough surfaces.

In order to further investigate the effect of surface roughness, a comprehensive SPRAY study was performed using both surface inputs based on artificially roughened microscopy images and computer generated structures. It was revealed that RMS is not a suitable parameter to assess the sputter yield of rough surfaces, since structures with...
same RMS value can have very different sputter yields. In contrast, $\delta$ appears to characterise rough surfaces reasonably well and allows to predict the sputter yield independent from the scale of roughness. Determination of $\delta$ is achievable with common microscopy techniques, while high image resolution was found to be important for consistent results. A major outcome was that under normal ion incidence, rough surfaces with $\delta < 20^\circ$ show sputter yields which can be approximated by the value for a perfectly flat surface under incidence angle $\varphi = \delta$. We furthermore presented an empirical fit formula, which only requires $\delta$, $\varphi$ and a sputter yield value under normal ion incidence for a flat tungsten surface as input parameters to describe the sputter yield for a wide range of ion incidence angles $\varphi$ and roughness parameters $\delta$. Although the presented results were only obtained for argon ions on tungsten, the SPRAY code just considers geometric effects. Therefore, we expect that similar trends can also be found for other ion-target combinations, even though the actual magnitude and angular dependence of the sputter yield are of course strongly material dependent. Key applications range from process optimisation in PVD techniques where the surface roughness of crucibles can alter deposition rates, to lifetime predictions in nuclear fusion devices. The revealed correlation between sputter yield and $\delta$ may also provide a good starting point for a more refined theoretical description of sputtering effects on rough surfaces in future studies.

CRediT authorship contribution statement


Acknowledgements

The authors are thankful for the continuous support with the QCM electronics by Michael Schmid (TU Wien). Sample manufacturing was in part performed by Leonard Raumann, Jan Willem Coenen (Forschungszentrum Jülich, Institute of Energy and Climate Research, Germany) and Martin Oberkofler (Max Planck Institute for Plasma Physics, Garching, Germany) and is highly acknowledged. The computational results presented have been achieved in part using the Vienna Scientific Cluster (VSC-3), project ID 70998.

This work has been carried out within the framework of the EUROfusion Consortium and has received funding from the Euratom research and training program 2014-2018 and 2019-2020 under Grant Agreement No. 633053. The views and opinions expressed herein do not necessarily reflect those of the European Commission. Financial support has also been provided by KKKÖ (commission for the coordination of fusion research in Austria at the Austrian Academy of Sciences - ÖAW). The authors acknowledge the TU Wien Bibliothek for financial support through its Open Access Funding Program.

Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.apusc.2021.151204.