Study of surface evolution via Modified Ion Beam Sputtering Semi-Empirical Model (MIBSEM)

Oluwole E. Oyewande\textsuperscript{1,2}, Samuel A. Awelewa\textsuperscript{1}, T.V. Omotoso\textsuperscript{3}

\textsuperscript{1}Department of Physics, University of Ibadan, Ibadan, Nigeria.
\textsuperscript{2}Department of Physics, College of Science & Technology, Covenant University, Ota, Nigeria.

oluwole.oyewande@covenantuniversity.edu.ng

Abstract. An Ion Beam Sputtering Monte Carlo Model (IBSM) was modified to study the effect of different surface diffusion mechanics on the surface height and surface roughness of materials when bombarded with energetic ions. The extension was done by incorporating Seahet al.’s semi-empirical ion sputtering equations into the sputter erosion algorithm of the MC model and by enforcing curvature dependence of sputter yield. Also, TRansport of Ions in Matter (TRIM) software, was used to calculate the sputter yield and the values obtained were compared with those obtained from Seahet al.’s semi-empirical model and the sputter yield obtained from sputtering experiments. Our studies show that the surface diffusion mechanics that occurs during a sputtering process controls the substrate surface height and its roughness while curvature dependence of sputter yield has little or no contribution to the target surface height and its roughness. Our comparison of the calculated sputter yield values of TRIM and Seahet al.’s model with experimental sputter yield values show that Seahet al.’s model gives sputter yield values closer to experimental values than TRIM at a normal angle of incidence while TRIM gives sputter yield values closer to experimental values at 60° angle of incidence.

Keywords: sputtering, sputter yield, surface pattern, Monte Carlo, TRIM.

1. Introduction
When an energetic ion strikes a solid surface, atoms are not only ejected from the solid (i.e. sputtering) but also the surface morphology of the target is altered in the process [1]. Interestingly, the resulting surface topography of the target could be a variety of self-assembled nanoscale patterns, including ripples, nanodots and arrays of sharply pointed conical protrusions, with sizes in the nanometer range [2]. Although a significant amount of research has been done towards understanding the mechanics responsible for pattern formation but there is lack of full comprehension of this phenomenon. Meanwhile, a good understanding of the physical mechanisms will make sputtering a simple, fast and relatively less expensive method for industrial production of nanomaterials.

Pattern formation by Ion Beam Sputtering (IBS) has been widely studied in three major theoretical frame works, namely: Molecular dynamics simulations in the classical physics frame work [3-4], Monte Carlo (MC) simulations in the statistical framework [5-10] and simulations with continuum models in a mathematical frame work of linear and nonlinear partial differential equation [11-13]. In this work, our study of pattern formation on surfaces bombarded with energetic ions was done in the
statistical frame work. One of the advantages of MC simulations is that it enables us to independently control various atomistic processes [9]. Thereby, we can study how each of these processes controls pattern formation behaviour.

A basic ion beam sputtering MC model consist of two major segments; erosion steps and surface diffusion steps. In must work done in this frameworkSigmund’s sputtering mechanics [14] have been employed in the modelling of sputtering erosion algorithm. However, Sigmund’s mechanism makes sputter yield at a valley higher than the sputter yield at a crest of self-assembled nanoscale patterns [9] and leads to high sputter yield compared to experimental yield [10]. We have made use of Seah et al.’s semi-empirical model to obtain the sputter yield used in [the sputtering erosion algorithm of our MC model.

This model ensures that our sputtering erosion algorithm simulates experimental erosion as much as possible, knowing that the formula gives a more accurate sputtering yield value compared to that given by Sigmund’s theory, because it is semi-empirical. Seah et al.’s model is a semi-empirical equation for the sputter yields of mono-elemental solids using neon, argon and xenon ions for the energy range of 250eV – 10,000eV [15, 16]. The equation is an improved version of Matsunami et al.’s [17] and Yamura and Tawara’s [18] semi-empirical formula.

Also, in most cases the works done on ripple pattern formation have been channelled towards studying the characteristics and properties such as wavelength and amplitude of the ripples formed [9-11]. But in this paper, we have studied the effect of different surface diffusion mechanisms, sputter re-deposition and curvature dependence of sputter yield on the surface of a sputtered target, using a semi-empirical Monte Carlo Model.

The rest of the paper is arranged as follows. Section 2 gives the explanation of how the existing IBSEM was extended for our study of surface evolution. In section 3 we present and discuss the results of the simulations with the extended model. Finally we present our conclusions in section 4.

2. Methodology

This chapter discusses the model used in this work for studying surface pattern formation by energetic ion beam. In this work, an Ion Beam Sputtering Semi-Empirical Model (IBSEM) [5] was improved on by incorporating Seah et al.’s semi-empirical model into IBSEM. A full explanation of how IBSEM was developed and works is given in Ref. 5. Also,TRansport of Ions in Matter (TRIM) software and Seah et al.’s model were separately used to generate the sputter yield values of silicon and tungsten. We then compared these sputtering yield values with experimental values obtained from Refs.19 and 20 so as to ensure that the sputter yield used in the model is close to experimental sputtering yield as much as possible.

The Seah et al.’s sputter yield semi-empirical model is given by equations 1 and 2, where \( Y(0) \) gives the value of the sputtered yield at a normal angle of incidence, \( U_o \) (in electron-volts) is the surface binding energy per atom of the target atom. \( E \) is the incident ion energy, \( E_{th} \) is the threshold energy for sputtering, \( s \) is a parameter which equals 3.8, \( S_n(E) \) is the nuclear stopping power per atom and \( \theta \) is the ion beam angle of incidence. The theoretical background of this semi-empirical model has been introduced in [15, 16]. In this work, equations 1 and 2

\[
Y(0) = \frac{0.042s\alpha}{U_o} S_n(E) \left[ 1 - \frac{E_{th}}{E} \right]^{1/2} \tag{1}
\]

\[
Y(\theta) = Y(0)(sec \theta)^f \exp[-f \cos \theta_{max} (sec \theta - 1)] \tag{2}
\]

were incorporated into the Ion Beam Surface Erosion Algorithm (IBSEA) of IBSEM to able to study surface pattern formation beyond sputtering at a normal angle of incidence and at angle 45° used in Ref. 5, since Seah et al.’s tables [21] only provide sputtering yield at the two angles mentioned. We also implemented curvature dependence of sputter yield in IBSEM. In addition to incorporating Seah et al.’s model into IBSEA, the model was also used to simulate sputtering yield of Ar⁺, Ne⁺ ions on silicon and tungsten at a normal angle of incidence and at 60° and the results obtained were compared
with those obtained from TRIM simulations and experimental results. This was done firstly to ensure that the sputter yield used in the extended IBSEM is close to experimental sputter yield as much as possible. Secondly to investigate if the sputter yield values from TRIM software could be used in Ion Beam Sputtering Model for the study of surface evolution of materials and incident energy range not covered by Seah et al.’s model. We limited our investigation to these two angles of incidence because we could only access experimental sputter yield values at these two angles. TRIM is a Monte Carlo computer program that calculates the interactions of energetic ions with amorphous targets [22]. It gives detailed data about all sputtered atoms and the sputter yield of the target [23]. Details of the theoretical background for the calculations and simulations embedded in TRIM package is discussed in the papers by Ziegler and Biersack [23]. In each of the TRIM simulations, we used the substrate thickness of 100nm and the default density of each substrate. Also, 1000 ions were used for each simulation and a detailed calculation with full damage cascade was performed for each simulation.

3. Result and Discussion
The results of our simulations are presented and discussed in this section. From the plots of the sputter yield obtained from experiments with that of TRIM and Seah et al.’s model shown in Fig. 1, it was noted that at a normal angle of incidence, Seah et al.’s model gives sputter yield values closer to experimental values than that of TRIM and vice versa at 60° angle of incidence. These results call for more investigation into how the accuracy of these two ( sputter yield from Seah et al.’s model and TRIM ) vary at other angles but due to lack of access to more experimental data, further research could not be done on this in this work. However, the results show that sputter yield generated with TRIM simulations could as well be used in IBSEA and in other MC sputtering models to study surface pattern formation by other incident ions not captured by Seah et al.’s model.

![Figure 1](https://example.com/figure1.png)

**Figure 1:** Comparison of sputter yield of Silicon for Ar⁺ ions at a Normal angle incidence and 60° from Experiment, TRIM and Seah et al.’s SEM. Experimental data were obtained from [19] and [20] respectively.
For all the results reported below, simulation time is in Monte Carlo steps (which represents incidence ion flux) and the surface average height and roughness are in lattice spacing, with order of lattice constant.

The extended model was used to simulate the sputtering of 1keV and 10keV Ar\(^+\) ion on silicon from 0°−89° angle of incidence. However the results of the average surface height and the surface roughness of the simulation of 1keV Ar\(^+\) on silicon at angle 80° are reported below.

The results for simulation with no Curvature Dependence Algorithm (n-CDA) and Surface Reorganization Algorithm (SRA) are shown in Fig. 2. The plot of the average surface height showed that it decrease with simulation time and the surface roughness plot showed that surface roughness increases with time. Simulations with Curvature Dependence Algorithm (CDA) and Surface Reorganization Algorithm gave same results as shown in Fig. 2.

The results for simulation with no Curvature Dependence Algorithm (n-CDA) and Surface Diffusion Algorithm (SDA) are shown in Fig. 3. The plot of the average surface height showed that it increases with simulation time and the surface maintains its smoothness for a simulation period of about 500 Monte Carlo steps but afterwards the surface begins to get roughen and the roughness keeps increasing with simulation time. Simulations with Curvature Dependence Algorithm (CDA) and Surface Diffusion Algorithm (SDA) gave the same results as shown in Fig. 3.

The results of the simulation with no Curvature Dependence Algorithm (n-CDA), Surface Diffusion Algorithm (SDA) and Surface Re-organization Algorithm (SRA) gave similar results as when CDA, SDA and SRA was used. (see Fig. 4). But there was a little drop in the value of the final average height and surface roughness. This shows that the implementation of curvature dependence of sputter yield and preventing shadowing effect leads to an increase in the surface height and surface roughness even though the contribution is insignificant.

![Figure 2: Simulation with 1keV Ar\(^+\) on Si at 80° with the implementation no CDA and SRA](image-url)
(a) Average height of surface profile with simulation time  
(b) Surface roughness as a function of simulation time

**Figure 3**: Simulation with 1keV Ar$^+$ on Si at 80° with the implementation no CDA and SDA

(a) Average height of surface profile with simulation time  
(b) Surface roughness as a function of simulation time

**Figure 4**: Simulation with 1keV Ar$^+$ on Si at 80° with the implementation no CDA, SDA and SRA
The results for the simulation with Curvature Dependence Algorithm (CDA), Surface Diffusion Algorithm (SDA) and Re-deposition Algorithm (RD) are shown in Fig. 5. The plot of the average surface height shows that it increases with simulation time and the surface maintains its smoothness for a simulation period of about 500 Monte Carlo steps but afterwards the surface begins to get roughen and the roughness keeps increasing with simulation time. Since there is no much difference between this simulation and that with CDA and SDA, this implies that re-deposition of sputtered atoms has little impact on the surface roughness by reducing it.

4. Another section of your paper
In this work, we aimed at using an extended Monte Carlo Semi-Empirical Model to study the surface evolution of solid materials by ion beam sputtering. Different simulations were performed with the sputtering process simulated using Sehat et al.’s semi-empirical model with the implementation of curvature dependence of sputter yield (and without the implementation), surface diffusion and surface re-organization of atoms. The effect of re-deposition of sputtered atoms was also considered.

We were able to show that the surface diffusion mechanics that takes place during the sputtering process is the major factor that controls the surface height and surface roughness of a sputtered material. And that curvature dependence of sputter yield and redeposition of sputtered atoms has little or no contribution to the surface height and surface roughness.

Also, our studies show that sputter yield from TRIM software could also be used in Ion Beam Erosion Algorithm of an Ion Beam Sputtering Monte Carlo Model.

References
[1] Tapobrata S. and Dinakar K. (2012). Nanofabrication by Ion-Beam Sputtering.
[2] Bradley MR, Redeposition of sputtered material is a nonlinear effect, Physical Review B 83, 075404(2011).
[3] Rosandi R, Maureen LN, and Herbert MU, Subsurface channeling of keV ions between graphene layers: Molecular dynamics simulation, Physical Review B, vol. 91, pp.
125441(2015).

[4] Herbert MU, Bradley RM, Maureen LN, and Woflhard M, Sputter yield of curved surfaces, Physical Review B, Vol. 91, pp. 165418(2015).

[5] Oyewande EO, A quasi-conserved particle monte carlo model of surface evolution with semi-empirical sputter yield modulated erosion: 1kev Ar$^+$ sputtering of si, Transactions of the Nigerian Association of Mathematical Physics(2015).

[6] Maureen LN, Luis S, Herbert MU, and Woflhand M, Sputtering of Si nanospheres, Phys. Rev. B 90, 045417(2014).

[7] Zhangcan Y, Michael AL, and Jean PA, Kinetic Monte Carlo simulation of self-organized pattern formation induced by ion beam sputtering using crater functions, Phys. Rev. B 91, 075427(2015).

[8] Oyewande EO, Alexander KH, and Reiner K, Propagation of Ripples in Monte Carlo Models of Sputter Induced Surface Morphology, Phys. Rev. B. Vol. 71, pp. 195405(2004)

[9] Chan WL and Chason E, Making waves: Kinetic processes controlling surface evolution during low energy ion sputtering. J. Appl. Phys. 101(2007).

[10] Hartmann AK and Kree R, Long-time effects in a simulation model of sputter erosion. Physical Review B, Volume 65, 193403(2002).

[11] Bradley RM and Harper JME, Theory of ripple topography induced by ion bombardment, J.Vac. Sci. Technology A, Vol.6, p. 2390.

[12] Cuerno R and Barabasi AL, Dynamic scaling of ion-sputtered surfaces, Phys. Rev. letters 74(23),4746(1995).

[13] Oyewande OE and Adeoti BB, Theory of off-normal incidence ion sputtering of surfaces of Type AxB1-x and a conformal map method for stochastic continuum models, African Rev. of Physics. Vol. 9, pp. 0024(2014).

[14] Sigmund P, Theory of Sputtering. I. Sputtering Yield of Amorphous and Polycrystalline Targets. Physical review volume 184, Number 2(1969).

[15] Seah MP, Clifford CA, Green FM, and Gilmore IS, An accurate semi-empirical equation for sputtering yields I: for argon ions. Surf. Interface Anal.2005; 37: 444-458.

[16] Seah MP, An accurate semi-empirical equation for sputtering yields II: for neon, argon, and xenon ions. Nucl. Instrum. Methods Phys. Rev. B, Vol. 229, pp 348-358(2005)

[17] Matsunami N, Yamamura Y, Hikawa Y, Itoh N, Kazumata Y, Miyagawa S, Morita K, Shimizu R, Tawara H, Energy Dependence of Sputtering Yields of Monoatomic solids. IPPJ-AM-14(1980).

[18] Yamamura Y and Tawara H, Energy Dependence of Ion-induced Sputtering Yields from Monoatomic Solids at Normal Incidence. At. Data Nucl. Data Tables 1996; 62: 149.

[19] Laegreid N and Wehner GK, Sputtering Yields of Metals for Ar$^+$ and Ne$^+$Ions with Energies from 50 to 600eV. J. Appl. Phys. 32, 365(1961).

[20] Gernot E, Rastislav K, Vladimir K, Yuri T, Pezoldt J, The estimation of sputtering yields for SiC and Si. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 196 (1-2), 39-50,2002.

[21] Seah MP, Clifford CA, Green FM, and Gilmore IS, Surface and Nanoanalysis: Sputter Yield Values. National Physical Laboratory. [online] August 29, 2014. http://www.npl.co.uk/science-technology/surface-and-nanoanalysis/services/sputter-yield-values.

[22] Ziegler JF, Biersack JP and Litmark U, The Stopping and Range of Ions in Solids,New York :Pergamon(1985).

[23] Ziegler JF, Ziegler MD and Biersack JP,The stopping and range of ions in matter, Nucl. Instrum. Meth. Phys. Res. B 268, 1818 – 1823(2010).