DOCTORAL THESIS

Predictive modelling of gas assisted electron and ion beam induced etching and deposition

A thesis submitted in fulfilment of the requirements
for the degree of Doctor of Philosophy

School of Physics and Advanced Materials
University of Technology Sydney

Author
Alan Stephen BAHM

Supervisors
Prof. Milos TOTH
Associate Prof. Michael FORD

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Certificate of Original Authorship

I, Alan Stephen BAHM, certify that the work in this thesis titled, 'Predictive modelling of gas assisted electron and ion beam induced etching and deposition' has not previously been submitted for a degree nor has it been submitted as part of requirements for a degree except as fully acknowledged within the text.

I also certify that the thesis has been written by me. Any help that I have received in my research work and the preparation of the thesis itself has been acknowledged. In addition, I certify that all information sources and literature used are indicated in the thesis.

Signature of Student:

Date: 28 July 2016

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Contributing Publications

Peer-reviewed publications that contributed to this work:

- Spontaneous Growth of Gallium-Filled Microcapillaries on Ion-Bombarded GaN, Aurelien Botman, **Alan Bahm**, Steven Randolph, Marcus Straw, and Milos Toth *Physical Review Letters* 111, 135503 - Published 25 September 2013
- Dynamic Pattern Formation in Electron-Beam-Induced Etching, Aiden A. Martin, **Alan Bahm**, James Bishop, Igor Aharonovich, and Milos Toth *Physical Review Letters* 115, 255501 - Published 18 December 2015

Non-Contributing Publications

Peer-reviewed publications not featured in this work containing research undertaken during the PhD program:

- Localized Probing of Gas Molecule Adsorption Energies and Desorption Attempt Frequencies, Jared Cullen, **Alan Bahm**, Charlene J. Lobo, Michael J. Ford, and Milos Toth, *Journal of Physical Chemistry C*, 2015, 119 (28), pp 15948-15953

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Abbreviations

API	Application Programming Interface
BCA	Binary Collision Algorithm
BET	Brunauer-Emmett-Teller theory
BDF	Backwards Difference Formula
CM	Centre of Mass
CPM	Closest Point Method
CPU	Central Processing Unit
EBIE	Electron Beam Induced Etching
EBIED	Electron Beam Induced Etching and Deposition
EDS	Energy Dispersive X-ray Spectroscopy
FEM	Finite Element Methods
FMM	Fast Marching Method
FVM	Finite Volume Methods
GIS	Gas Injection System
GPU	Graphics Processing Unit
FIB	Focused Ion Beam
HJE	Hamilton-Jacobi Equation
iSE	Ion Generated Secondary Electrons
ISM	Ion-Solid-Modelling
ITK	Insight Segmentation and registration Toolkit
IBIED	Ion Beam Induced Etching and Deposition
LLF	Local-Lax-Friedrichs
LF	Lax-Friedrichs

LSM	Level Set Method
MC	Monte Carlo
MD	Molecular Dynamics
MTL	Mass Transport Limited
PDE	Partial Differential Equation
RD	Reaction Diffusion
SEM	Scanning Electron Microscope
SIMD	Single Instruction Multiple Data
SRIM	Stopping and Range of Ions in Matter (software program)
TEM	Transmission Electron Microscope
TRIM	Transport and Range of Ions in Matter (software program)
TRIDYN	Transport of Ions (Dynamic)
UV	Ultraviolet
1D	One Dimensional
2D	Two Dimensional
3D	Three Dimensional

Physical Constants

Bohr radius $a_0 = 5.291\ 772\ 109... \times 10^{-11}$ m
Electron rest mass $m_e = 9.109\ 382\ 15... \times 10^{-31}$ kg
Vacuum permittivity $\epsilon_0 = 8.854\ 187\ 817... \times 10^{-12}$ F/m

Pi $\pi = 3.14159...$
Euler's number $\mathrm{e} = 2.71828...$

Symbols

R^3	real coordinate space of three dimensions
∇	gradient
∇^2 or Δ	Laplacian a.k.a. Laplace operator
∇_S	intrinsic gradient
∇_S^2 or Δ_S	intrinsic Laplacian a.k.a. Laplace-Beltrami operator
Δ	discrete difference
∂x	partial derivative of x
dx	total derivative of x
$t \to 0$	“as t goes to zero”
$\boldsymbol{x}, \boldsymbol{v}, \boldsymbol{n}, \boldsymbol{t}$	vector position, velocity, normal, tangent
κ	curvature
ϕ_t	time derivative of ϕ
$\hat{H}$	exact Hamiltonian
H	discretized approximation of the Hamiltonian
max	maximum
$O(N)$	big O notation
$F_{ext}(\phi = 0) = F$	“extension velocity at $\phi = 0$ is F”
log	logarithm base 10
ln	natural logarithm
$\approx$	approximately equal to
$\equiv$	definition
cos, tan, cot	cosine, tangent, cotangent function
$cp()$	closest point operator

Abstract

While the field of experimental micrometre scale EBIED / IBIED ("electron beam chemistry" or "ion beam chemistry") has been growing in recent years, the 3D simulation of these systems at real scales has been non-existent. This type of simulation is important for it is only in three dimensions that interesting asymmetric and patterning phenomena can be tracked.

There are a couple of difficulties in these types of simulations. One is solving the diffusion of adsorbate concentrations in the system. Accurate simulation of diffusion on general 2D surfaces is non-trivial, (even on 1D curves), and can require unnatural re-parametrization of the surface (re-meshing). Another difficulty is that simulations have generally been atomistic and limited in scale. The key to providing large scale 3D simulations comes from applying new, mathematically robust, computer-science methods based on implicit surfaces to this field.

In this thesis, the issues above are addressed in a couple of different ways. In one case, diffusion over a complex surface was reduced to piecewise axially symmetric equations. Later, implicit methods for solving adsorbate kinetics continuum equations and evolving the surface are implemented, the closest point method and the level set method respectively. The development of the tools themselves is a non-trivial exercise as there are few software libraries for the level set method and none for the closest point method. These tools were then used independently to simulate etching and diffusion, as well as in concert to demonstrate the ability to simulate 3D deposition in the mass transport limited and reaction rate limited regimes.