INVESTIGATIONS OF SHEATH FORMATION DURING PLASMA-SURFACE INTERACTION

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by

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Dedicated to

my family and loved ones

&

those

whoever inspired and encouraged me

Certificate

This is to certify that the thesis entitled "**Investigations of Sheath Formation during Plasma-Surface Interaction**" being submitted by **Mr. Rajat Dhawan** is worthy of consideration for the award of the degree of Doctor of Philosophy and is a record of the original bonafide research work carried out by him under my guidance and supervision, and that the results contained in it have not been submitted in part or full to any other university or institute for award of any degree / diploma.

I certify that he has pursued the prescribed course of research. I approve the thesis for the award of the degree of Doctor of Philosophy.

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RDhawan

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Abstract

In microelectronics, semiconductors and automobiles industries, the surface properties of the adopted materials degrade continuously and also their applications are being constrained because of the issues of poor corrosion resistance, wear resistance, surface roughness, fatigue failure, and lower hardness. The performance of a material is determined by its surface properties. Therefore, in order to explore engineering industries in a greater detail, it is necessary to minimize hostile environments so that the material surfaces can be protected. One of the imperative ways to deal with such problems is the plasma-based material processing through the understanding of plasma-surface interactions. Plasma-based surface treatments are vitally used to modify the specific surface biological property while bulk properties remain unaltered. In order to understand the mechanism of plasma-surface interactions, it is necessary to understand the concept of sheath, which is a thin layer of charged species that is formed surrounding a metallic conducting surface when it comes into the contact of a plasma.

Electronegative plasma is composed of electrons, negative ions and positive ions. Such plasmas are being widely adopted for the last two decades because of their expanding applications in many fields such as spacecraft propulsion, microelectronics industries, thin-film deposition and sputtering, mass spectroscopy, plasma-based surface processing, and many more. Such plasmas are preferred for these applications because of relatively less impact of electrons on the surface of the material under investigation. In many applications, they serve as ion source because of the requirement of both positive and negative ion beams; these have also been used in low energy beam applications and also when energetic electrons can produce destructive effects on the surface. The best method to ward off irregular shapes is the addition of negative ions to counterbalance the positive ions, assemble on the wafer and also to enhance the working of materials using in the fabrication of integrated circuits. Electronegative plasmas are widely adopted for soft substrates to have a defect-free analysis, as they develop immensely small sheath voltage in comparison with electropositive one.

A large number of theoretical models have been developed to study sheath formation criterion and behaviour of plasma parameters. However, these models still have some limitations. For example, in most of the investigations, the researchers assumed the behaviour of negative ions through their Boltzmann distribution, which is far from the reality due to the neglect of their mass. Since the difference of mobilities of the positive and negative ions is not much and it is not appropriate to consider Boltzmann distribution for only negative ions, both

these ions should be treated at equal footings with inclusion of their masses. Moreover, the mass-ratio of the ions is also neglected, which plays a crucial role in the sheath formation process during plasma-based material processing. A very few mathematical models are available in the literature which investigate the influence of non-extensive distributed electrons on the sheath characteristics. Therefore, it is desirable to develop new models to study the realistic situation in a greater detail.

In the present thesis, keeping in mind all the above points, we have developed theoretical models where the finite mass of ions is considered and the behaviour of both the positive and negative ions is taken to be governed by the fluid equations. We have also considered the drift term in the momentum transfer equation of the negative ions to explore the practical case by taking into account the generalized situation of their different masses from the positive ions. In view of the realistic situation, the cases of presence and absence of the negative ions' drift term has been compared and it is shown that this drift term has a significant effect on the sheath formation criterion. The impact of negative-to-positive ion mass-ratio on the behaviour of plasma parameters and sheath characteristics has also been analyzed. We have established a relation which speaks about the sheath thickness. Our mathematical models have explained the exact sheath thickness determination and plasma parameters, like potential profile, the density of the charged species, ions velocity, etc. in the sheath region. The case of doubly charged ions is also considered herewith. A mathematical model has also been developed to investigate two-temperature non-extensive distributed electrons in the electropositive warm plasma having finite collisions and ionizations. Some singularities were encountered in the mathematical treatment, which have been discussed in detail and are removed while applying appropriate boundary conditions. One of the important results is the absence of oscillatory structure of the potential when the negative ions are described by their fluid equations (as that of the positive ions), instead of their Boltzmann distribution as assumed by the other workers. In magnetized collisional electronegative warm plasma if the magnetic field is strong, then the profile of positive-ion density near the sheath edge has shown a pulse-like structure for the case when we neglect the components of the positive ions' velocity parallel to the probe/wall at the sheath edge. This pulse-like structure disappeared when all the components of the positive ions velocity at the sheath edge have been included.

The outcome of this thesis would be advantageous in the experiments like plasma-based material processing where higher potential gradient is preferred to have a better thin films formation, since the positive ions are accumulated within a short region and results in preferred thin film formed. These results would help experimentalists to select a particular plasma for a specific application. These results would help to understand those experiments where the negative ions are intentionally added to counterbalance the positive ions to ward off irregular shapes. Such results would enhance the working of materials used in the fabrication of integrated circuits. It is also expected to play a significant role in understanding astrophysical plasmas and plasma reactors at low-pressure conditions, where two types of the electrons are formed and the density distribution of these species is far from their usual Boltzmann distribution. The problem of communication blackout, which is occurred when a hypersonic speed is attained by space vehicles during their travelling through the atmosphere, can also be addressed.

सार

माइक्रोइलेक्ट्रॉनिक, अर्धचालक और ऑटोमोबाइल उद्योगों में अपनाये गए पदार्थों की सतह के गुणों में लगातार गिरावट आती है और खराब संक्षारण प्रतिरोध, धातु श्रांतिकी प्रतिरोध, सतह खुरदरापन, श्रांतिज पात और कम कठोरता के मुद्दों के कारण उनके अनुप्रयोगों को भी बाधित किया जा रहा है। किसी पदार्थ का प्रदर्शन उसकी सतह के गुणों से निर्धारित होता है। इसलिए, इंजीनियरिंग उद्योगों का अधिक विस्तार से पता लगाने के लिए, प्रतिकूल वातावरण को कम करना आवश्यक है ताकि पदार्थों की सतहों को संरक्षित किया जा सके। ऐसी समस्याओं से निपटने के अनिवार्य तरीकों में से एक प्लाज्मा-सतह की अन्योन्यक्रिया की समझ के माध्यम से प्लाज्मा-आधारित सामग्री प्रसंस्करण है। प्लाज्मा आधारित सतह के उपचार का उपयोग विशिष्ट सतह जैविक गुण को संशोधित करने के लिए किया जाता है जबकि बल्क गुण अपरिवर्तित रहते हैं। प्लाज्मा-सतह के अंतःक्रियाओं के तंत्र को समझने के लिए, म्यान की अवधारणा को समझना आवश्यक है, जो कि आवेशित प्रजातियों की एक तनु परत होती है जो एक प्लाज्मा के संपर्क में आने पर धातु की संवाहक सतह के आसपास बनती है।

इलेक्ट्रोनगेटिव प्लाज्मा इलेक्ट्रॉनों, नकारात्मक आयनों और सकारात्मक आयनों से बना होता है। इस तरह के प्लाज़्मा को पिछले दो दशकों से अंतरिक्ष यान प्रणोदन, माइक्रोइलेक्ट्रॉनिक उद्योग, तनु-फिल्म जमाव और स्पटरिंग, मास स्पेक्ट्रोस्कोपी, प्लाज्मा-आधारित सतह प्रसंस्करण, और कई अन्य क्षेत्रों में उनके विस्तारित अनुप्रयोगों के कारण व्यापक रूप से अपनाया जा रहा है । जांच के तहत पदार्थ की सतह पर इलेक्ट्रॉनों के अपेक्षाकृत कम प्रभाव के कारण इन अनुप्रयोगों के लिए ऐसे प्लाज़्मा को प्राथमिकता दी जाती है। कई अनुप्रयोगों में, वे सकारात्मक और नकारात्मक आयन बीम दोनों की आवश्यकता के कारण आयन स्रोत के रूप में कार्य करते हैं; इनका उपयोग कम ऊर्जा बीम अनुप्रयोगों में भी किया गया है और तब भी जब ऊर्जावान इलेक्ट्रॉन सतह पर विनाशकारी प्रभाव पैदा कर सकते हैं। अनियमित आकार को दूर करने का सबसे अच्छा तरीका वेफर पर इकट्ठा हुए सकारात्मक आयनों को संतुलित करने के लिए नकारात्मक आयनों को जोड़ना है, और एकीकृत सर्किट के निर्माण में उपयोग की जाने वाली सामग्रियों के काम को बढ़ाने के लिए भी है। इलेक्ट्रोनगेटिव प्लाज़्मा को व्यापक रूप से नरम सब्सट्रेट के लिए एक दोष-मुक्त विश्लेषण के लिए अपनाया जाता है, क्योंकि वे इलेक्ट्रोपोसिटिव प्लाज़्मा की तुलना में अत्यधिक छोटे म्यान वोल्टेज विकसित करते हैं।

म्यान निर्माण मानदंड और प्लाज्मा मापदंडों के व्यवहार का अध्ययन करने के लिए बड़ी संख्या में सैद्धांतिक मॉडल विकसित किए गए हैं। हालाँकि, इन मॉडलों की अभी भी कुछ परिसीमानताएँ हैं। उदाहरण के लिए, अधिकांश जांचों में, शोधकर्ताओं ने बोल्ट्जमान वितरण के माध्यम से नकारात्मक आयनों के व्यवहार को अपनाया है, जो कि उनके द्रव्यमान की उपेक्षा के कारण वास्तविकता से बहुत दूर है। चूँकि धनात्मक और ऋणात्मक आयनों की गतिशीलता का अंतर अधिक नहीं है और केवल ऋणात्मक आयनों के लिए बोल्ट्जमान वितरण को अपनाना उचित नहीं है, इन दोनों आयनों को उनके द्रव्यमानों को शामिल करते हुए समान स्तर पर व्यवहार किया जाना चाहिए। इसके अलावा, आयनों के द्रव्यमान-अनुपात की भी उपेक्षा की जाती है, जो प्लाज्मा-आधारित सामग्री प्रसंस्करण के दौरान म्यान निर्माण प्रक्रिया में महत्वपूर्ण भूमिका निभाता है। साहित्य में बहुत कम गणितीय मॉडल उपलब्ध हैं जो म्यान विशेषताओं पर गैर-व्यापक वितरित इलेक्ट्रॉनों के प्रभाव की जांच करते हैं। इसलिए, यथार्थवादी स्थिति का अधिक विस्तार से अध्ययन करने के लिए नए मॉडल विकसित करना वांछनीय है।

वर्तमान थीसिस में, उपरोक्त सभी बिंदुओं को ध्यान में रखते हुए, हमने सैद्धांतिक मॉडल विकसित किए हैं जहां आयनों के परिमित द्रव्यमान पर विचार किया गया है और सकारात्मक और नकारात्मक

दोनों आयनों के व्यवहार को द्रव समीकरणों द्वारा नियंत्रित किया गया है। हमने सकारात्मक आयनों से नकारात्मक आयनों के भिन्न द्रव्यमानों की सामान्यीकृत स्थिति को ध्यान में रखते हुए व्यावहारिक मामले का पता लगाने के लिए नकारात्मक आयनों के गति हस्तांतरण समीकरण में बहाव शब्द पर भी विचार किया है। वास्तविक स्थिति को देखते हुए ऋणात्मक आयनों की प्रवाह अवधि की उपस्थिति और अनुपस्थिति के मामलों की तुलना की गई है और यह दिखाया गया है कि इस प्रवाह शब्द का म्यान गठन मानदंड पर महत्वपूर्ण प्रभाव पड़ता है। प्लाज्मा मापदंडों और म्यान विशेषताओं के व्यवहार पर नकारात्मक-से-सकारात्मक आयन द्रव्यमान-अनुपात के प्रभाव का भी विश्लेषण किया गया है। हमने एक संबंध स्थापित किया है जो म्यान की मोटाई के बारे में बताता है। हमारे गणितीय मॉडल ने म्यान क्षेत्र में सटीक म्यान मोटाई निर्धारण और प्लाज्मा मापदंडों, जैसे संभाव्यता प्रोफ़ाइल, आवेशित प्रजातियों का घनत्व, आयनों का वेग, आदि की व्याख्या की है। दोगुने आवेशित आयनों के मामले पर भी इसके साथ विचार किया गया है। परिमित टकराव और आयनीकरण वाले इलेक्ट्रोपोसिटिव गर्म प्लाज्मा में दो-तापमान गैर-व्यापक वितरित इलेक्ट्रॉनों की जांच के लिए एक गणितीय मॉडल भी विकसित किया गया है। गणितीय उपचार में कुछ विलक्षणताओं का सामना करना पड़ा, जिन पर विस्तार से चर्चा की गई है और उपयुक्त सीमा शर्तों को लागू करते हुए हटा दिए गए हैं। महत्वपूर्ण परिणामों में से एक संभाव्यता की दोलन संरचना की अनुपस्थिति है जब नकारात्मक आयनों को उनके बोल्टज़मैन वितरण के बजाय जैसा कि अन्य श्रमिकों द्वारा माना गया है उनके तरल समीकरणों (सकारात्मक आयनों की तरह) द्वारा वर्णित किया जाता है। मैग्नेटाइज्ड कोलिजनल इलेक्ट्रोनगेटिव वार्म प्लाज़्मा में यदि चुंबकीय क्षेत्र मजबूत है, तो म्यान किनारे के पास सकारात्मक-आयन घनत्व के प्रोफाइल में एक पल्स जैसी संरचना दिखाई देती है जब हम म्यान किनारे पर जांच/दीवार के समानांतर धनात्मक आयनों के वेग के घटकों की उपेक्षा करते हैं। यह पल्स जैसी संरचना गायब हो जाती है जब म्यान के किनारे पर सकारात्मक आयनों के वेग के सभी घटकों को शामिल किया गया।

इस थीसिस का परिणाम प्लाज्मा-आधारित पदार्थ प्रसंस्करण जैसे प्रयोगों में लाभदायक होगा, जहां अपेक्षाकृत अच्छी तनु फिल्मों के निर्माण के लिए उच्च संभाव्यता ढाल को प्राथमिकता दी जाती है, क्योंकि सकारात्मक आयन एक छोटे क्षेत्र के भीतर जमा होते हैं और इसके परिणामस्वरूप वांछित तनु फिल्म बनती है। ये परिणाम प्रयोगवादियों को एक विशिष्ट अनुप्रयोग के लिए एक विशेष प्लाज्मा का चयन करने में मदद करेंगे। ये परिणाम उन प्रयोगों को समझने में मदद करेंगे जहां अनियमित आकार को दूर करने के लिए सकारात्मक आयनों को संतुलित करने के लिए जानबूझकर नकारात्मक आयनों को जोड़ा जाता है। इस तरह के परिणाम एकीकृत परिपथों के निर्माण में उपयोग की जाने वाले पदार्थों के कामकाज में वृद्धि करेंगे। इन परिणामों से एस्ट्रोफिजिकल प्लाज़्मा और कम दबाव की स्थिति में प्लाज़्मा रिएक्टरों को समझने में भी महत्वपूर्ण भूमिका निभाने की उम्मीद है, जहां दो प्रकार के इलेक्ट्रॉन बनते हैं और इन प्रजातियों का घनत्व वितरण उनके सामान्य बोल्ट्ज़मान वितरण से बहुत दूर है। संचार ब्लैकआउट की समस्या, जो तब होती है जब अंतरिक्ष वाहनों द्वारा वातावरण के माध्यम से यात्रा के दौरान एक अतिध्वनिक गति प्राप्त की जाती है, को भी संबोधित किया जा सकता है।

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List of Symbols

α = Normalized collisional parameter

α_c = Background concentration ratio of cold electrons to total electrons

α_h = Background concentration ratio of hot electrons to total electrons

ε_0 = Permittivity of the free space

C = Polytropic constant

c_s = Sound/acoustic speed

δ = Ionization parameter/non-neutrality parameter

$\vec{\nabla} P_N$ = Pressure gradient term for negative ions

$\vec{\nabla} P_P$ = Pressure gradient term for positive ions

e = Electronic charge

$\vec{F_c}$ = Collisional drag force

γ = Power factor or dimensionless factor ranging from -1 to 0

γ_c = Normalized cold electrons temperature

γ_h = Normalized hot electrons temperature

γ_N = Normalized negative ions temperature

γ_P = Normalized positive ions temperature

i_N = Negative ion current collected by probe

i_P = Positive ion current collected by probe

j_N = Negative ion current density

j_P = Positive ion current density

K_B = Boltzmann constant

K = Geometric constant

L = Finite plasma size/characteristics length of plasma

λ_D = Debye length

Λ = Ionization length

M_e = Mass of electrons

M_N = Mass of negative ions

M_P = Mass of positive ions

μ = Mass ratio of electrons to positive ions

n_c = Cold electrons density

n_e = Electron density

n_{e0} = Electron density in plasma/background density of electrons

n_g = Neutral gas density

n_h = Hot electrons density

n_N = Negative ions density

n_{N0} = Negative ion density in plasma/background density of negative ions

n_P = Positive ions density

n_{P0} = Positive ion density in plasma/background density of positive ions

N_c = Normalized cold electrons density

N_e = Normalized electron density

N_h = Normalized hot electrons density

N_N = Normalized negative ion density

N_{Net} = Net space charge density

N_{N0} = Electronegativity

N_P = Normalized positive ion density

N_{P0} = Electropositivity

υ = Collisional frequency

υ_{att} = Attachment frequency

υ_{det} = Detachment frequency

υ_{iz} = Ionization frequency

v_N = Speed/velocity of negative ions

v_{N0} = Negative ion velocity at sheath edge

v_P = Speed/velocity of positive ions

ϕ = Electric potential

ϕ_{wall} or ϕ_P = Electric potential at wall/probe

ψ = Normalized electric potential

ψ' = Normalized electric field

ψ_{wall} or ψ_P = Normalized potential at the wall/probe

ψ_0 or ψ_s = Normalized potential at sheath edge

ψ'_0 or ψ'_s = Normalized electric field at sheath edge

q = Non-extensivity of the system

r = Distance from the surface of spherical/cylindrical probe

$\sigma(v)$ = Momentum-transfer collisional cross-section between the ions and the neutrals

T_c = Temperature of cold electrons

T_e = Temperature of electrons

T_{eff} = Effective temperature

T_h = Temperature of hot electrons

T_N = Temperature of negative ions

T_P = Temperature of positive ions

U_N = Normalized negative ion velocity

U_{N0} = Normalized negative ion velocity at sheath edge

U_P = Normalized positive ion velocity

U_{P0} = Normalized positive ion velocity at sheath edge

$U_{P_{Probe}}$ = Positive ion velocity at probe/wall surface

v_{P0} = Positive ion velocity at sheath edge/positive ion drift velocity at x = 0

x = Distance

z = Space coordinate in pre-sheath scale

ξ = Normalized distance

ξ_{PP} = Normalized position of probe/wall

ξ_s = Normalized distance corresponding to sheath edge

Z_N = Charge on negative ions

Z_P = Charge on positive ions

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