COMPARISON OF HIGH POWER IMPULSE MAGNETRON SPUTTERING AND MODULATED PULSED POWER SPUTTERING FOR INTERCONNECT METALLIZATION

BY

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DISSEPTION

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Abstract

Ionized physical vapor deposition is used to deposit the barrier and seed layers in the state of the art interlevel metallization process. As the critical dimension keeps shrinking, it has become increasingly difficult to use the current techniques to form thin, continuous and stable barrier/seed layers, and more likely to form void during the following process of electroplating. An enhanced metal ionization is believed to be critical. Ions in the deposition flux can increase the nucleation density and adhesion of Cu to Ta due to their surface penetration, and create less overhang due to the high directionality of ion flux.

In this work, high power impulse magnetron sputtering (HiPIMS) and its derivative, modulated pulsed power (MPP) magnetron sputtering, with their claimed high ionization capability, are proposed for the application of barrier/seed layer deposition. Their plasma properties and metal ionization fractions are characterized using various pulsing and discharge parameters. Depositions on patterned wafers are performed to evaluate their potential for interconnect metallization application. Time- and spatially-resolved plasma diagnostics are further performed to investigate the physical mechanisms involved in the pulsed plasma generation, evolution, and plasma transport. Specially designed experiments and plasma modeling are used to further understand some key features during HiPIMS, such as the self-sputtering process.

Fundamental studies of HiPIMS discharge are conducted in a planar magnetron. Very high peak current up to 750 A can be achieved. Triple Langmuir probe (TLP) is adopted to measure the electron density \( n_e \) and electron temperature \( T_e \). High electron densities \( n_e \) during the pulse to about \( 5 \times 10^{17} \text{ m}^{-3} \) are measured on the substrate and reach \( 3 \times 10^{18} \text{ m}^{-3} \) later after the pulse ends. Cu ionization fractions (IF) are measured on the substrate level using a gridded energy analyzer (GEA) combined with a quartz crystal microbalance (QCM). Up to 60% has been achieved using a 200 Gauss magnetic field configuration, much higher than the DC magnetron sputtering. It basically increases with higher charging voltage and longer pulse length due to higher plasma densities. However, lower ion extraction efficiency at stronger B field, however, leads to lower Cu ionization in spite of a higher
plasma density.

HiPIMS has been shown to have some complicated and distinctive features. Their possible effects on the application, as well as the underlying physics are investigated. Plasma expansion is observed with a high plasma density peak moving from the target to the substrate. It has varied speed and preferred orientation. Different parameters such as the charging voltage, pulse duration, and magnetic field strength are found to affect the plasma transport. A large plasma potential drop is observed in the presheath and extends into the bulk plasma region during HiPIMS discharge. It not only affects the plasma expansion but also determines the ion extraction efficiency, which is critical for the interconnect metallization application. A direct evidence of the self-sputtering effect is provided by measuring the incident fluxes to the cathode through a hole in the target. Plasma is initially ignited only in a long strip in the race track where the B field is strong and drifts toward the weak-B region. High fraction of Cu\(^+\) flux is determined.

To provide more insights into the development of Cu ion and Ar ion species, a time-dependent model is built to describe the ionization region where plasma is confined by magnetic field. The important processes such as plasma-target interactions, electron collision ionizations, and gas rarefaction are incorporated in the model. The test results of the model show the capability to predict the temporal development of the electron density, the degrees of ionization for Cu and Ar, and the ratio of Cu\(^+\) ions to Ar\(^+\) ions.

Magnetic field configurations are modified specifically for the HIPIMS. The race track pattern is varied to optimize the target utilization and the downstream plasma uniformity. A closed path for electrons to drift along is found essential in the design. The configuration of wider race track generates a higher pulse current, and extends the intense plasma coverage on the substrate. A spiral-shaped magnetic field configuration is able to generate high pulse current, achieve a downstream plasma with superior uniformity, and yield a better target utilization even without the assistance of magnet rotation.
Modulated pulsed power (MPP) magnetron sputtering is a new derivative of the HiPIMS that may allow unprecedented user control over the growth process. It has some distinctive features, such as flexible control over the discharge voltage and current waveforms. In this study, a thorough characterization of the MPP discharge using two different models (Solo and Cyprium) is performed in the Galaxy planar magnetron to better understand this pulsing technique. The effects of various pulsing and discharge parameters, as well as the magnetic field, are studied.

For the test of deposition on patterned wafers, a hollow cathode magnetron is chosen. All three types of power supplies, DC, MPP and HiPIMS are first subject to the plasma characterization, both to study the discharge mechanisms on HCM and to develop potentially good recipes with high Cu ionization fractions. Both MPP and HiPIMS increase the Cu ionization fraction in the deposition flux (up to 25% and up to 30% respectively) as compared with the normal DC magnetron sputtering (as below 20%). Ultimately, Cu is deposited on patterned wafers with trenches of different widths as narrow as 70 nm. The conformality of the Cu film on the trench will be compared using cross-section scanning electron microscopy (SEM). Reduced overhang is achieved using MPP Solo as compared with DC sputtering. More significant improvements have been seen using the MPP Cyprium model. HiPIMS also shows slightly better conformality and may be further improved with appropriate substrate biasing. The potential of applying HiPIMS and MPP for barrier/seed layer deposition will be further discussed.
To my family
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Chapter 1  Introduction

The semiconductor industry has been devoted to constantly improving the performance of computer chips. Computers are seen to have greater capabilities while cost less than those from just a few years ago. The improvements are the result of an increase in the number of transistors that can be fit into a given area. Moore's Law [1] predicted years ago that the number of transistors on integrated circuits doubles approximately every two years. This trend has proven to be surprisingly accurate, in part because the law is now used in the semiconductor industry to guide long-term planning and to set targets for research and development.

Dictated by Moore's Law, the device dimensions keep shrinking to below 22 nm as of 2013. Enormous challenges are encountered during many fabrication steps used to build the integrated circuit (IC). These include the interlevel metallization process. To interconnect the various metal layers and the semiconductor devices, Cu lines are inlaid in the previously etched trenches and vias in the dielectric layers [2]. One crucial step to achieve reliable interconnect metallization is to form high quality and conformal Ta(N) barrier and Cu seed layers before Cu electroplating is performed [3,4]. However, using the current employed physical vapor deposition (PVD) techniques, it has become increasingly difficult to form thin, continuous and stable barrier/seed layers, and more likely to form void during electroplating.

An enhanced metal ionization is believed to be beneficial [5]. A larger amount of Cu ions in the deposition flux, after being accelerated by the wafer sheath potential, can increase the nucleation density and adhesion of Cu to Ta due to their surface penetration. The high directionality of ion flux also leads to much less overhang built up at the top edge of the trenches or vias as well as better step coverage.

Different methods have been developed to enhance the metal ionization, as the so-called ionized physical vapor deposition (iPVD) techniques [6-8]. The methods usually involve an enhanced magnetic field confinement to increase the plasma density as in the hollow cathode magnetron (HCM) [9,10], or a secondary high density plasma for further ionizing the sputtered atoms [11,12]. Unfortunately at the current device node, even these techniques are reaching their limit. New method
to further extend the PVD deposition is urgently wanted, especially when other techniques such as chemical vapor deposition (CVD) and atomic layer deposition (ALD) are not suitable for high-quality Cu deposition.

High power impulse magnetron sputtering (HiPIMS) is a relatively new concept. It was developed in 1990s [13] and has since been extensively investigated for functional coatings. In HiPIMS, very high voltages (around 1 kV) are applied in short pulses to the target. It leads to quite high pulse power densities of several kW/cm². Two or three orders of magnitude denser plasmas than the DC magnetron plasmas and high degrees of ionization have been observed, for example 70% for Cu [8,13]. This renders HiPIMS a promising alternative for the barrier/seed deposition. However, very little has been done for such a study, likely because the fast-paced and cost-driven semiconductor industry usually shows less interest in other techniques unless the mainstream techniques face extreme difficulty for further improvement.

In order to develop suitable HiPIMS processes for the interconnect metallization, a better understanding of the HiPIMS discharge is necessary. There have been great efforts to characterize the HiPIMS plasmas and explore the discharge mechanisms in the past decade. The pulsed plasmas are revealed to have complex and unique behaviors. However, the underlying physics in HiPIMS are far from being fully understood and an effective control over various plasma properties is still difficult.

Other than the conventional HiPIMS, some new derivatives have been invented such as the modulated pulsed power (MPP) technique [14-16]. MPP discharge allows customization of the discharge voltage and current waveforms and thus flexible user control over the growth process. It has also been claimed to have high metal ionization without significant deposition rate loss as observed in HiPIMS. This MPP technique hasn’t been widely adopted and a more thorough study is still needed.

Motivated by all the above necessities, this research will be focused on characterizing the plasma properties and behaviors during HiPIMS and MPP discharge, further exploring and understanding the unique physical mechanisms, and developing HiPIMS and MPP processes to evaluate their potential for interconnect metallization application.
Chapter 2  Overview

2.1  Background

2.1.1  Interlevel Metallization in IC Fabrication

The state of the art interlevel metallization process in Very-large-scale integration (VLSI) manufacturing employs the so-called damascene process where Cu lines are inlayed in the previously etched trenches and vias in the dielectric layers. [2] The lines are formed by electroplating over barrier and seed layers typically formed by metal sputtering process which is also referred to as physical vapor deposition (PVD). The most common barrier consists of TaN/Ta and Cu is used as the seed layer. Figure 2.1 illustrates the involved steps including the dielectric deposition, patterning by lithography, etching, stripping, barrier/seed layer deposition (step I and J), electrofill, chemical mechanical polishing (CMP), etc.

![Dual damascene process for the interlevel metallization](image)

The reliability of the interconnect metallization depends on the quality of different interfaces,
especially on the barrier/seed interface [3,4]. Failure to ensure continuous seed coverage inside the recessed features with good adhesion to the barrier may result in voids during electroplating, post-CMP defects, stress migration, and electromigration failures. Several factors which are inherent to PVD process complicate the task. The neutral deposition flux has a wide angular distribution resulting in deposition build up on the feature corners called overhang, which shadows the sidewall leading to the poor step coverage. Thin sidewall film tends to be discontinuous due to island formation during the film growth [17]. Ta and Cu are immiscible metals and do not form alloys at any temperature. This makes Ta a good diffusion barrier for Cu. The same time the absence of chemical bonds makes the wetting of Cu on Ta difficult [18]. The rough resulting film can be easily attacked by electrolyte during initiation phase in the plating bath leading to the seed dissolution and subsequent plating voids.

The key factor in building a stable film is the presence of energetic metal species in the deposition flux. It had been demonstrated that Cu forms a much stronger interface with the barrier in the case of sputter deposited films compared to evaporated films [19]. Sputtered Cu atoms leave the target with relatively high energy but lose it rapidly due to collisions with the background gas [20]. The fraction of Cu$^+$ ions in the deposition flux is usually relatively low, but it is very important. While their energy in the plasma is similar to the energy of neutrals, ions are accelerated by the wafer sheath potential. Thus presence of Cu$^+$ ions can increase the nucleation density and adhesion of Cu to the Ta due to their surface penetration. The direction of sheath electrical field results in high directionality of ion flux, therefore ions also have much better step coverage ability. Due to all these reasons, a higher fraction of ions in depositing flux is highly desirable, leading to the development of different methods to enhance ionization, i.e. the ionized physical vapor deposition (iPVD) techniques.

### 2.1.2 Magnetron Sputtering

Magnetron sputtering is one of the most important physical vapor deposition (PVD) techniques. It allows deposition of a large number of metal and compound coatings with specified mechanical, electrical and optical properties, and has been used for a wide field of coating applications.
Figure 2.2 A planar magnetron arrangement to create a static magnetic field parallel to the target surface, where secondary electrons are retained and drift in the $E \times B$ direction following a cycloidal path.

In a DC magnetron sputtering (dcMS) process, ionized inert gas atoms are accelerated by a DC negative bias applied to the cathode (target). The ion bombardments cause the target atoms to be ejected, or sputtered out, and then condensate on a substrate to form a film. A magnetron is designed to magnetically enhance the plasma by adding permanent magnets behind the target, as shown in Figure 2.2. Magnetic field is created to be parallel to the target surface and perpendicular to the electric field, so that electrons are trapped in a ring-shaped region in the vicinity of the target due to the $E \times B$ drift. The high plasma density enhances the ionization process and thus increases the sputtering rate locally to form a “racetrack”. [21]

Magnetic field design is important in DC magnetron sputtering (dcMS) to affect the plasma and the film deposition [22-25]. The strength of B field determines the efficiency of plasma confinement and thus the sputtering rate. The B field profile on the target surface defines the drifting path of electrons and the resultant erosion race track. It is generally desirable to have a full face target erosion to control the re-deposition and to extend the target lifetime. For a sputtering process, it is usually critical to have uniform downstream plasmas and deposition rates, which are affected by the race track pattern as well as the degree of unbalancing in the magnetic field [22, 25]. The magnetic field configuration has to be carefully customized and combined with magnet pack rotation or scanning to optimize the magnetron performance, especially for deposition on large substrates.

One of the main advantages of magnetron sputtering over the other PVD techniques such as evaporation is the additional kinetic energy of the plasma species, including both the neutral sputtered
atoms and a certain amount of ionized species. These species impinge onto the film and transfer energy to the adatoms. As a result, the surface and bulk diffusion processes are enhanced, allowing tailoring of structural, optical, electrical, and mechanical properties of the films. [26-28]

2.1.3 Ionized Physical Vapor Deposition

Ion species in a magnetron sputtering are especially interested since their energy can be controlled independent of the flux, by applying a bias to the substrate. Moreover, the ion flux is directional, which is beneficial for applications such as barrier and seed layers deposition in high-aspect-ratio trenches and vias. Unfortunately, in the DC magnetron sputtering processes the degree of ionization of the plasma particles is relatively low [29]. Methods to produce a higher fraction of ions in depositing flux are desirable.

Ionized physical vapor deposition (iPVD) has been developed as a high plasma density tool to increase the ion flux. An iPVD essentially consists following steps: 1) creating a metal vapor flux by physical methods, such as the sputtering and evaporation; 2) ionizing the metal neutral using a high density secondary plasma source or a single enhanced ionization source; 3) collimating the ion flux by the plasma sheath or negative bias before deposition [7,8]. A typical iPVD system involves a DC (or RF) magnetron, and a secondary high density plasma, which is commonly an inductively coupled plasma (ICP) or an electron cyclotron resonance (ECR) plasma [30-33]. Utilizing ICP plasma up to 40% of Cu atoms in the sputtered flux can be ionized, but they have to be slowed down through thermalization for effective ionization [8]. The remaining 60% of the deposition flux is then comprised of slow neutrals, similar to those in evaporating systems. In some systems, a single source can be used for both sputtering and ionizing the target material, such as the hollow cathode magnetron (HCM) [9,10].

The iPVD-based processes have proved viable for liner seed through 45 nm node [34]. However, as the critical dimension of features keeps decreasing, it's more and more difficult for iPVD to keep up with the demand of high metal ion fraction in the deposition flux.
2.1.4 High Power Impulse Magnetron Sputtering (HiPIMS)

Other than the above-mentioned iPVD techniques based on the application of a secondary discharge to create a dense plasma, which more or less complicates the system, another concept to achieve dense plasma has been investigated in parallel since 1990s, by using a pulsed mode [13,35,36]. For a typical magnetron, the plasma can be intensified simply by increasing voltage and thus power, until it reaches an upper limit that the target is overheated. To avoid overheating, voltage are applied to the target as pulses with low duty ratios (<10%) and frequencies (<10 kHz) to maintain a low average power. Meanwhile, the high pulsed voltages (around 1 kV) lead to quite high pulse power densities of several kW/cm², producing high power impulse magnetron sputtering (HiPIMS) [8,13]. A typical waveform of the pulse voltage and current is shown in Figure 2.3. To be clarified, HiPIMS used in this thesis always doesn’t include modulated pulsed power (MPP) technique as described in the next section.

![Typical waveform of pulse voltage and current](image)

**Figure 2.3** A typical voltage and current time trace for the high power pulsed discharge. The pulse was 100 µs long. A Cu cathode was used and the sputtering gas, Ar, was maintained at 0.065 Pa (0.5 mTorr) (data taken from [13]).

There have been numerous studies on different aspects of HiPIMS discharge, which were well reviewed in Refs [8,37,38]. High peak electron densities (n_e) during pulses in the order of $10^{18}$-$10^{19}$ m⁻³ were observed in the substrate vicinity [39-41] as compared to less than $10^{16}$ m⁻³ in the
conventional DC magnetron sputtering [29]. The intense HiPIMS plasma not only enhanced the ionization of gas atoms, but also greatly increased the degree of ionization of the sputtered material, as measured by deposition rate change on floating and biased substrates [13,42] and by spectroscopic analyses [42-49]. Some examples were 70% for Cu [13], 30% for Cr [50], 9.5% for Al [51], and over 90% for Ti [43]. The significant fraction of metal ions promoted a “self-sputtering process” that the pulsed plasma evolved from Ar ion dominated to metal ion dominated [42,52]. This mechanism was also believed to account for a loss of deposition rate as compared with dcMS under a same average power [44,52]. This is one main drawback of HiPIMS since it was developed.

By using HiPIMS, high ion fluxes are made available at the substrate. The magnitude and the composition of these fluxes could be varied by changing the process parameters [38,41,42,44,52]. HiPIMS has been applied for metal and compound film deposition and modification, to achieve an ultra-dense structure and a very smooth surface, to tailor the phase, to modify electrical and optical properties of the films, or to enhance the film adhesion by ion implantation [37,53], etc. It has also been successfully used for coating complex-shaped substrates [54].

### 2.1.5 Modulated Pulsed Power (MPP) Magnetron Sputtering

Modulated pulsed power (MPP) technique is a relatively new method of applying the basic principles of HiPIMS [14,15,16]. One feature of the MPP technique is that it generates long pulses (500 to 3000 μs instead of 100 μs in HiPIMS), whose shape can be arbitrarily adjusted. As shown in Figure 2.4, each of these long pulses (macropulses) is composed of a train of “micropulses”, typically 20-40 μs each. By modulating the on- and off-time (τ_{on} and τ_{off}) of micropulses, as well as the charging voltage and repetition frequency of the macropulse (10 to 400 Hz), customized voltage and current waveforms can be generated. More degrees of freedom are thus offered for additional process control during sputtering.
MPP can provide a high peak current throughout the macropulse, and as a result, high plasma density and ionization fraction were observed [16,55,56]. MPP technique has been used to fabricate dense and uniform films [16,55]. The loss of deposition rate in MPP discharge as compared with dcMS was shown to be less significant than that in a HiPIMS discharge [16].

2.2 Past Research Studies on HiPIMS

2.2.1 Plasma Characterization

A high enough electron density is necessary to promote the electron impact ionization during the discharge. The information of electron density is commonly obtained using a single Langmuir probe. For temporal studies of the HiPIMS plasma, a single probe was usually combined with a data acquisition system for data sampling at different time delays [57]. By varying the voltage applied to the single probe, a set of probe I-V traces at different time in and after a pulse could be re-constructed and analyzed to get the electron density \( n_e \). It was shown that the peak electron density in HiPIMS was in the order of \( 10^{18}-10^{19} \) m\(^{-3}\) in the substrate vicinity [39-41], about two or three orders of magnitude higher than the density in a conventional dcMS. Alami et al. [41] measured the temporal behavior of the plasma density while varying the process conditions, as shown in Figure 2.5. They observed a second peak after the pulse had been switched off and attributed it to an ion acoustic wave reflecting off the chamber wall back to the measurement position. Gylfason et al. and Alami et al.
stated that such a shock solitary ion acoustic wave was generated soon after the plasma ignition due to the large plasma impedance gradient between the pulse on-time and off-time [58,41].

Figure 2.5 Temporal and spatial variations of the electron saturation current $I_e$ in a HiPIMS discharge with a Ta target, varying (a) the sputtering gas, (b) the chamber radius, (c) the probe location, and (d) the pulse power (from Alami et al. [41]).

The Langmuir probe measurements could also be used to achieve the time-dependent electron energy distribution function (EEDF) during pulses. Gudmundsson et al. [40,59] showed a transition
from a broad energy distribution in the early stage of the pulses to a double Maxwellian distribution toward the end of the pulse, and finally a Maxwellian-like distribution hundreds of µs after the pulse had been switched off, as shown in Figure 2.6. The depletion in the high energy tail was believed to be caused by the escape of high energy electrons to the chamber walls and inelastic collisions of high energy electrons. An obvious cooling of the electrons was observed as the effective electron temperature ($T_{\text{eff}}$) dropped from 1.5-2 eV early in the pulse to 0.3-0.7 eV at the end of the pulse and later. Such effective electron temperatures are actually lower than what were observed in a conventional dc magnetron sputtering discharge in the range of 2-4 eV. The decreased $T_{\text{eff}}$ during a HiPIMS pulse was due to that electron impact excitation and ionization of the metal atoms have much lower excitation thresholds and ionization potential than the argon gas.

Other than the single Langmuir probe, double probe [60] and triple probe [41,61] were also found useful for temporal characterization of the pulsed plasma. For example, the triple Langmuir probe allows a direct display of the electron density $n_e$ and temperature $T_e$, without the need of applying a swiping voltage on the probe. More details on the building and operation of a triple probe as well as the based theory are explained in the next chapter.

Figure 2.6 The EEPF (electron energy probability function) for an HiPIMS plasma with a 100 µs on-time and a 50 Hz frequency at 1.33 Pa (10 mTorr) (from Gudmundsson et al. [40]).
Spectroscopic analyses, such as optical emission spectroscopy (OES), absorption spectroscopy, and mass spectroscopy are commonly employed to differentiate different ion species and neutrals. The OES studies showed much higher metal ion emission intensity in HiPIMS than in dcMS [42], and an increased ion species emission as the peak target current density increased as shown in Figure 2.7 [52]. Quantification of the ion population are done mainly by using weight gain differences on a floating and a positively biased substrate [13,42], absorption spectroscopy [44,46,47] and mass spectroscopy [48,49]. Large degrees of ionization for metals ranging from about 10% to over 90% were determined for different target materials, e.g., 70% for Cu [13], 30% for Cr [50], 9.5% for Al [51], and over 90% for Ti [43]. Both singly and doubly charged metal ions were observed [42,48]. For example, a highly metallic plasma consisted of 50% Ti⁺, 24% Ti²⁺, 23% Ar⁺, and 3% Ar²⁺ during HiPIMS [48]. It should be noted that many reported results of the ionization degrees, are inconsistent among different research groups.

Figure 2.7 Temporal optical emission spectroscopy under increased peak target current density during high power impulse magnetron sputtering. An increase of the Cr⁺/Cr⁰ ratio indicated an increased
ionization degree. Ar$^+$ and Ar$^0$ emission intensities decrease as a result of rarefaction in (a) - (d) (data taken from [52]).

Both time-averaged and time-resolved ion energy distribution functions (IEDFs) were measured using mass spectrometer combined with the electrostatic gating of ions [48,62,63]. According to the study in Ref [63], IEDF of Ar$^+$ is peaked at 3 eV and spreads up to 20 eV. Ti$^+$ counts in the energy distribution have a maximum at 22 eV while their energies extend up to 100 eV. The reason for high ion energy observed for the HiPIMS discharge is at present time not fully understood.

### 2.2.2 HiPIMS Discharge Mechanisms

The plasma diagnostics revealed that the HiPIMS plasma and its temporal evolution are quite complex. The plasma also induces some very unique features in HiPIMS discharge. One distinctive feature of the HiPIMS is the self-sputtering, i.e., target being sputtered by ionized target materials. Anders et al. [64] showed an example of HiPIMS sputtering of Al (Figure 2.8). Above a certain voltage limit, the target current was sustained at a high level throughout the pulse indicating a sustained self-sputtering. The effects of self-sputtering were observed to become dominate at a high-enough peak current [44,52,64]. This is likely originated from the significant fraction of metal ions in the plasma, and the gas atoms being depleted in the target vicinity by the high flux of sputtered materials as the so-called rarefaction phenomenon [65], as confirmed by OES in Figure 2.7. Anders et al. [66] also suggested that multiply charge particles play an important role to enhance the self-sputtering since these impinging particles generate the secondary electron emission more efficiently.
Figure 2.8 Current pulse shapes at different constant target voltages for Ar–Al HiPIMS discharges. Above a certain voltage limit, the target current is sustained at a high level throughout the pulse indicating a self-sustained self-sputtering [64].

Other than the self-sputtering effect, the HiPIMS plasma has been observed to have another unique phenomenon, as the strong localization of ionization [67-69]. As shown in Figure 2.9, the plasma over a magnetron’s erosion “racetrack” is not azimuthally uniform but concentrated in distinct dense ionization zones which move in the E×B direction with about 10% of the electron E×B drift velocity. The phenomena are proposed to be caused by an ionization instability where each dense plasma zone exhibits a high stopping power for drifting high energy electrons, thereby enhancing itself. Ionization zones move because ions are “evacuated” by the electric field. Therefore, later arriving electrons drift further along the racetrack until they find particles to interact with.
Loss of deposition rate is another topic being extensively studied, considering it is an obstacle towards the industrialization of HiPIMS. The HiPIMS deposition rates were found 10-40% of those in dcMS, depending on the target material [7], as shown in Figure 2.10. Christie [70] developed a model showing that the decrease of HiPIMS deposition rate is related to the enhanced ionization of the sputtered material, and/or the re-direction of ionized sputtered species to the target, as well as lower
self-sputtering yields than Ar$^+$ sputtering yields. Experiments further confirmed that high ionization and Ar rarefaction are necessary conditions for triggering the self-sputtering and simultaneous loss of the deposition rate [52,44].

Figure 2.10 Deposition efficiency for HiPIMS and conventional dcMS plotted for several metal targets: relative deposition rate vs. self-sputtering yield $S_S$ divided with its Ar-sputter yield $S_{Ar}$ (from Helmersson et al. [7]).

Despite the extensive studies on the electron and ion fluxes, the transport of the pulsed plasma was usually only qualitatively explained. As opposed to the theory of ion acoustic wave [58], Bohlmark et al. [71] proposed an anomalous transport of electrons across the magnetic field lines and a simultaneous deformation of the magnetic field occur in HiPIMS instead of obeying the classical theory of plasma diffusion. Meanwhile, ions are subject to a tangentially outward force due to the azimuthal electron current above the race track, which reduces the deposition rate [72]. It is argued that the localization of ionization zones also induces abnormal electron diffusion. Each region of strong azimuthal plasma density gradient generates an azimuthal electric field, which promotes the escape of magnetized electrons and the formation of electron jets and plasma flares [67,68].

### 2.3 Remaining Concerns

After a brief review of the past research studies on the HiPIMS plasma characterization and discharge mechanisms, it can be seen that the underlying physics in HiPIMS is far from being fully
understood and an effective control over various plasma properties is still difficult.

First, there are inconsistencies in the plasma diagnostic results (e.g. ionization degree, electron temperature, plasma diffusion rate) among different research groups. The underlying reason may be the different plasma generators, vacuum chambers, operation conditions, and diagnostic tools being used. It will be beneficial to conduct an investigation using an independent system and cross-check some basic properties and features with the previous HiPIMS studies. Different diagnostic tools such as triple Langmuir probe and gridded energy analyzer combined with QCM (as explained in Figure 3.12) can be used as to compare with the primarily used single Langmuir probe, OES and mass spectroscopy, etc.

Second, the mechanisms during the HiPIMS are still not well understood. Many observations, such as the plasma transport during and after the pulses and the evolution of the plasma density and electron/ion energy distribution, still lack solid explanations. Also, despite various results indicating the rarefaction and self-sputtering effects in HiPIMS, there hasn’t been any direct measurement of the fluxes to the cathode to confirm them.

Third, the optimization of magnetic field has been largely overlooked in HIPIMS. In most cases, rotating or scanning magnets designed for dcMS are commonly used. However, the behaviors of the pulsed plasma including the plasma ignition, the plasma growth, and the downstream plasma release are different from those in dcMS. The influence of the magnetic field configuration on these processes is undoubtedly important, but has not been systematically studied except for a few discussions regarding the magnetic field deformation in HIPIMS [48] or effect of magnetic field strength in modulated pulsed power magnetron sputtering [73].

Fourth, there are some preliminary investigations on the MPP sputtering showing a reasonably high ionization and a less loss of deposition rate. But a more thorough study of MPP is yet to be performed to find its advantages and disadvantages as compared with HiPIMS.

Last, the HiPIMS has been extensively used to deposit functional coatings but not much has been done to apply it for the potential application of barrier/seed layer deposition for interconnect metallization, which faces great challenges now.
2.4 Objectives

2.4.1 Thesis Statement

In this research, high power impulse magnetron sputtering (HiPIMS) and its derivative, modulated pulsed power (MPP) magnetron sputtering, will be compared in large commercial systems. Time- and spatially-resolved studies of the HiPIMS and MPP plasma properties and fluxes are performed to investigate their dependence on various discharge parameters. An innovative method to measure the fluxes through the plasma sheath to the cathode is developed. The unique but still unclear mechanisms involved in the plasma ignition and evolution, self-sputtering, and plasma transport, etc. are further investigated. Both HiPIMS and MPP will be evaluated for their potential for interconnect metallization application. Specific objectives are expanded upon this statement in the next.

2.4.2 Proposed Work

1. Study of HiPIMS and MPP plasmas

One of the objectives of this work is to perform a systematic comparison between HiPIMS and MPP sputtering and further explore their discharge mechanisms. The pulsed plasmas are characterized and compared in large commercial-size magnetrons, which have rarely been reported. Triple Langmuir probe is used to study the temporal plasma behaviors, such as the plasma generation and evolution. A detailed three-dimensional plasma diagnostics in the chamber is performed to study the plasma transport. This will contribute to improving the uniformity of the plasma and ultimately the film deposition.

2. HiPIMS and MPP for interconnect metallization

The potential of HiPIMS and MPP techniques for barrier/seed layers deposition will be studied. One critical concern is whether metal ions can be efficiently extracted from the pulsed plasma to the substrate instead of being returned to the cathode for the self-sputtering. Diagnostics are thus designed to measure the fraction of metal ions in the deposition flux. Based on the plasma diagnostics, processing recipes will be developed and optimized. Cu is then deposited in narrow trenches (70nm) by HiPIMS and MPP sputtering. The conformality of the deposition will be compared with the dcMS
and evaluated.

3. **Study of self-sputtering in HiPIMS**

Self-sputtering is believed to be a crucial feature in HiPIMS, as indicated by the OES measurements. In this work, experiments will be designed to provide direct characterization of self-sputtering by measuring fluxes of the incident species onto the target surface without disturbing the plasma. A special setup is designed with a small orifice drilled on the target. The incident plasma including Ar neutral and ions, metal neutral and ions and electrons can penetrate the orifice and reach the other side of the target. Mesh grid, current collecting plate, QCM, and witness Si wafers are used in combination for the differentiation of these species.

4. **Optimization of Magnetic Field Configuration**

The effect of magnetic field strength and field configuration on the HiPIMS and MPP discharge will be investigated. The plasma generation and transport will be characterized using the 3-D Langmuir probe measurements. New designs of magnetron configurations will be made to improve the target utilization and the uniformity of the downstream plasma.

5. **Plasma model for HiPIMS**

Plasma model can be useful to understand the physical mechanisms during HiPIMS discharge. A time-resolved model to study the ionization region will be built, by incorporating important processes such as plasma-target interactions and ionization process in plasma. Featured behaviors of the HiPIMS discharge will be depicted and important plasma properties such as electron density, metal ionization degree and ratio of metal ions to argon ions will be predicted. The model results will be compared with the experimental data.
Chapter 3  Experimental Setup

In this chapter, the employed magnetron systems and various diagnostic tools are introduced. Pulsed plasmas are studied on a planar magnetron with adjustable magnets assembly, and a hollow cathode magnetron. The latter is also used for trench deposition. Two different types of pulsed plasma generators are adopted, one for conventional HiPIMS and one for MPP sputtering. For diagnostics, single Langmuir probe and triple Langmuir probe are used to characterize DC plasmas and pulsed plasmas respectively. The fractions of metal ions in the deposition fluxes are determined using a gridded energy analyzer (GEA) combined with a quartz crystal microbalance (QCM). A special design including a small orifice on the target and biased grids and QCM behind the orifice was used to measure the fluxes towards the target.

3.1  Planar Magnetron System

3.1.1 Galaxy Planar Magnetron Chamber

A planar magnetron is very commonly used in various applications. It also has relatively simpler magnetic field configuration, e.g. than hollow cathode magnetron, to make it suitable for fundamental studies of pulsed magnetron discharge. An MRC Galaxy planar magnetron is therefore

Figure 3.1 MRC Galaxy planar magnetron sputtering tool.

A planar magnetron is very commonly used in various applications. It also has relatively simpler magnetic field configuration, e.g. than hollow cathode magnetron, to make it suitable for fundamental studies of pulsed magnetron discharge. An MRC Galaxy planar magnetron is therefore
employed. It is a commercial tool featuring 14 inch (35.6 cm) diameter circular planar target, as shown in Figure 3.1. The schematic diagram of the chamber is given in Figure 3.2. The target can be titanium (Ti), aluminum (Al), or copper (Cu), which is water-cooled to allow high power discharge. A rotatable magnets assembly is mounted behind the target. The pedestal height can be adjusted but is typically fixed at about 14 cm from the target surface. A turbo pump is equipped to achieve a base pressure lower than $5 \times 10^{-4}$ Pa. Pure argon (Ar) gas is supplied and controlled with a mass flow controller to get working pressures ranging from 1 to 15 mTorr ($1.3 \times 10^{-1}$ to 2.0 Pa). The pressure is monitored with a capacitance manometer.

![Figure 3.2 Schematic diagram of the Galaxy chamber with 3D triple Langmuir probe system.](image)

In this work, the effect of magnetic field configuration on HiPIMS discharge is subjected to study. The original magnets assembly in the Galaxy system was designed for DC magnetron sputtering (dcMS). It uses asymmetric arrangement of magnets as shown in Figure 3.3 (a), generating a horseshoe-shaped B field. The detailed B field profile is measured and shown in Figure 3.4 (a). Rotation of the magnet is needed to even out target erosion and deposition on the substrate. Other than this original magnet, a freely adjustable magnet pack is designed and built (Figure 3.3 b). Magnets can be placed into different slots in the stainless steel base plate to achieve varied magnetic field configurations.
3.1.2 Magnetic Field Configurations

Magnetic field configuration is one of the core designs in a magnetron. It affects the plasma intensity based on the degree of electron confinement. It is generally desirable to have a full face target erosion to control the re-deposition and to extend the target lifetime. For a sputtering process, it is usually critical to have uniform downstream plasmas and deposition rates, which are affected by the race track pattern as well as the degree of unbalancing in the magnetic field [22, 25]. The magnetic field configuration has to be carefully customized and combined with magnet pack rotation or scanning to optimize the magnetron performance, especially for deposition on large substrates. The old Galaxy magnet pack, for example, has an asymmetric horseshoe-shaped B field as shown in Figure 3.4 (a). A long race track is formed by connecting the outer and the inner circles. With rotation, other than the inner and outer circular erosion groves, the area in between is also sputtered.

To study the effect of B field effect in HiPIMS discharge, COMSOL is used to assist the magnetic field design by calculating the magnetic flux intensities in the three-dimensional discharge space for any proposed arrangement of magnets. $B_\parallel$, as the component parallel to target, is determined on the target surface. Its maximum value is used here to represent the magnetic field strength. The shape of $B_\parallel$ resembles the probable shape of the race track, providing a good approach for the race track pattern design. The COMSOL calculation has been verified by comparing with the experimental
magnetic field measurement. Figure 3.4 (b) shows the magnet arrangement mimicking the old Galaxy magnetic field configuration. It has a long race track but slightly lower $B_y$ with a maximum of about 320 Gauss. The configuration will be referred as “300GL” standing for about 300 Gauss and long race track.

![Old magnet design](image1)

![300GL design](image2)

**Figure 3.4** (a) Old Galaxy magnet and the corresponding $B_y$ measured on the target surface. (b) A similarly-shaped B field configuration, 300GL, created using the adjustable magnet pack.

More designs are then made, starting with different B field strength. Two rows of magnets are kept at a same distance to yield a simple circular shape of race track, as illustrated in Figure 3.5. The numbers of magnets are varied in COMSOL to achieve the desired $B_y$ on the target surface, i.e. maximum at 200, 500, and 800 Gauss respectively. The simulation results of the B field are calibrated with the experimental measurements using a magnetometer. The three configurations will be referred as “200G”, “500G”, and “800G” in this thesis. More designs are shown in Chapter 6, where they are...
compared for HiPIMS discharge.

Figure 3.5 The magnet arrangements (top row) and the corresponding $B_y$ on the target surface (bottom row) in different configurations, (a) 200G, (b) 500G, (c) 800G, and (d) 500G-wide.

3.2 Hollow Cathode Magnetron

A 200-mm INOVA hollow cathode magnetron (HCM) commercial tool has been set up [10]. Figure 3.6 shows the schematic diagram of the system. The hollow cathode at the top consists of a copper (Cu) target in a shape of an inverted bucket. Other targets such as tantalum (Ta) and titanium (Ti) are also available. It enhances the electron confinement due to the hollow cathode effect, i.e., electrons can only escape through the bottom opening of the inverted-bucket-shaped target. One of the critical distinguishing characteristics of the source is its magnetic field. It exists not only in the source area, as in usual planar magnetrons, but also in the wafer area [74]. For unbalanced magnetrons in which the outer magnetic pole is stronger, there is a magnetic field separatrix between the target and substrate. It prevents electron loss and result in high density plasma confinement within the hollow cathode. The boundary separates these two areas, allowing to a certain extent independent manipulation of the sputtering plasma at the target and the depositing plasma at the wafer level.
normal magnetron effect combined with the hollow cathode effect and special magnetic field confinement enables the HCM to be a high-density iPVD system [75]. At the same power, the HCM exhibits plasma density one to two orders of magnitude higher than that of conventional planar magnetrons. [9,10]

The INVOA HCM chamber is about 673 mm in height and 381 mm in inner diameter as shown in Figure 3.6. The pedestal is movable in z direction and is typically set at 114 mm higher than the bottom surface of the chamber. A 200 mm diameter dummy wafer is placed on the pedestal during discharge. Both a cryopump and a turbopump are used to evacuate the chamber to a base pressure of $10^{-7}$ Torr. The target is sputtered using Ar gas with pressures between 1 and 15 mTorr. The input power for the HCM source is up to 32 kW during operation with the target well protected with water cooling.

A cylindrical Langmuir probe is first evaluated and then used to characterize the plasma parameters essential for understanding the HCM as a deposition tool. The probe is attached to a linear-rotatable feedthrough and can scan the region from the substrate level to 140 mm above, and from the center to the edge.

Figure 3.6 Schematic diagram of the 200mm INOVA hollow cathode magnetron system, showing (left) the magnetic field design inside the chamber and (right) the basic dimensions of the chamber.
3.3 Pulsed Plasma Generators

3.3.1 Huettinger HiPIMS Plasma Generator

A typical HiPIMS type generator, Huettinger TruPlasma Highpulse 4002 DC Generator, is used to power the magnetron. The plasma generator charges its capacitor bank up to 2000 V and outputs pulses of 1 to 200 µs long. A peak current up to 1 kA is allowed. The repetition frequency of pulsing is between 1 and 200 Hz. The charging voltage $V_{ch}$, the pulse duration $t_p$, and the repetition frequency $f$ are the basic pulsing parameters to control (Table 3.1). In the following sections, a set of HiPIMS discharge parameters is usually written in a form like 800 V, 50 µs, 100 Hz, 5 mTorr or 0.67 Pa without specifically mentioning the parameter names.

Table 3.1 Main pulsing parameters of the Huettinger pulsed plasma generator.

<table>
<thead>
<tr>
<th>Pulsing parameters</th>
<th>Output range</th>
</tr>
</thead>
<tbody>
<tr>
<td>Charging voltage ($V_{ch}$)</td>
<td>Maximum 4x500 V</td>
</tr>
<tr>
<td>Pulse voltage ($V_p$)</td>
<td>Maximum 4x500 V</td>
</tr>
<tr>
<td>Pulse current ($I_p$)</td>
<td>Maximum 1 kA</td>
</tr>
<tr>
<td>Mean power (P)</td>
<td>Maximum 10 kW</td>
</tr>
<tr>
<td>Frequency (f)</td>
<td>2 - 500Hz</td>
</tr>
<tr>
<td>Pulse duration ($t_p$)</td>
<td>1 - 200µs</td>
</tr>
</tbody>
</table>

Figure 3.7 Typical HiPIMS discharge voltage and current waveforms at different charging voltage.

The waveforms of the pulse voltage and current were measured using a high voltage probe (PMK PHV 661-L, 100:1 attenuation) and a Pearson current monitor (model 110, 0.1 V/A)
respectively. A typical set of discharge voltage and current waveforms are shown in Figure 3.7. These discharges are conducted in Galaxy planar magnetron using the 300GL magnetic field configuration and Al target. The parameters are 500-1000 V, 50 µs, 100 Hz, 5 mTorr.

3.3.2 **Zpulser MPP Plasma Generators**

Power can also be supplied to the magnetron by a Zpulser MPP Solo plasma generator. More information on the circuitry, specifications and operation of this MPP generator can be found in Refs [14, 15]. This plasma generator allows longer pulse durations (up to 3000 µs) than HiPIMS does. Each of these long pulses (called as “macro-pulse”) is composed of a sequence of “micro-pulses” (typically 20-30 µs in duration). By adjusting the “on” time (\(\tau_{on}\)) and “off” time (\(\tau_{off}\)) of the micro-pulses, macro-pulse width (500 to 3000 µs), and repetition frequency (10 to 400 Hz) using the Zpulser operating software, a macro-pulse of custom voltage and current waveform can be formed. The basic pulsing parameters used for Zpulser MPP plasma generators are given in Table 3.2.

<table>
<thead>
<tr>
<th>Pulsing parameters</th>
<th>Output range</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean power</td>
<td>0 - 10 kW</td>
</tr>
<tr>
<td>Peak power</td>
<td>0 - 147 kW</td>
</tr>
<tr>
<td>Pulse current ((I_p))</td>
<td>10 - 550 A</td>
</tr>
<tr>
<td>Mean power ((P))</td>
<td>0 - 10 kW</td>
</tr>
<tr>
<td>Frequency ((f))</td>
<td>10 - 400 Hz</td>
</tr>
<tr>
<td>Macro pulse duration</td>
<td>50 - 3000 µs</td>
</tr>
<tr>
<td>Mircopulse off time (\tau_{off})</td>
<td>6 - 40 µs</td>
</tr>
<tr>
<td>Mircopulse on time (\tau_{on})</td>
<td>2 - 18 µs</td>
</tr>
<tr>
<td>Duty cycle</td>
<td>Up to 28%</td>
</tr>
</tbody>
</table>

Multi-stage pulses are usually formed and applied to the cathode. Low voltage and current are used for the initial step to initiate the plasma with reduced arcing effect, and then by increasing the pulse voltage and current, a high-ionization stage can be reached [15]. A typical set of discharge voltage and current waveforms is shown in Figure 3.8 (a). It should be
clarified that the output voltage for each micro-pulse is not directly controllable, but varies with the $\tau_{on}$ and $\tau_{off}$. The voltage will increase as $\tau_{on}$ increases and $\tau_{off}$ decreases [14]. The present MPP plasma generator can deliver a peak power up to 147 kW, a maximum average target power of 10 kW, and a maximum peak current of 550 A to achieve a strong ionization. The discharge voltage, current, and power were monitored by the Zpulser operating software.

The power supply is later modified into a different Cyprium model. In this operation mode, the pulse profile is similar to that in Solo mode, with a macro-pulse composed of micro-pulses. But the circuitry is changed to allow the pulse voltage to quickly decrease in each micro-pulse off-time period. The concept is that if the voltage can drop to zero, then ideally the ions generated during on-time can be more efficiently extracted from the cathode to the substrate and benefit the deposition. The typical discharge IV characteristics are shown in Figure 3.8 (b). Note the pulse profile used is different from the one used in Figure 3.8 (a).

Both voltage and current exhibit obvious oscillations, although don’t usually reduce to zero as expected. Because of the oscillations, the power determination is less straightforward and is not output instantaneously from the power supply.

![Figure 3.8 Typical MPP discharge voltage, current and power waveforms in (a) Solo operation mode, (b) Cyprium mode. Note the difference in scales.](image)
3.4 Diagnostics

Multiple tools are used to characterize the plasma. Single Langmuir probes is constructed and used to determine the electron temperature and density. Triple Langmuir probes are used to determine the temporal evolution of the electron temperature and density. A gridded energy analyzer and a quartz crystal microbalance (QCM) are used in tandem to read total deposition rate and the neutral deposition rate in the magnetron system. A special diagnostic assembly is also built to measure the fluxes through a small hole on the target. These tools in combination provide a complete physical picture of what is going on inside of the different vacuum systems in this work.

3.4.1 Single Langmuir Probe

A Langmuir probe is the simplest and most commonly used tool to determine the principal parameters of plasmas such as the electron density \( n_e \), temperature \( T_e \), floating and plasma potential \( V_f \) and \( V_p \), and even the electron energy distribution function (EEDF). The interpretation of the data from it can be quite complicated though. It works by inserting one electrode into a plasma. By changing the voltage on the probe and measuring the current collected, a complete I-V trace can be acquired, as shown in Figure 3.9. The theory of the single Langmuir probe has been well reviewed in [76,77]. The practical method used for the probe analysis is described in Ruzic’s AVS chapter [78].

![Figure 3.9 A typical single Langmuir probe IV trace. The ion current, floating potential, and plasma potential are determined for the electron density and temperature calculation.](image_url)
The probe electronics are shown schematically in Figure 3.10. The Langmuir probe is biased via an electrical feedthrough. The probe signal passes through a BNC cable to the oscilloscope. The probe driving circuit consists of a BK PRECISION 4011A 5 MHz Function Generator, a Kepco Bipolar Operational Power Supply/Amplifier (Model BOP 500M), a Tektronix TDS 2014 4-Channel Digital Storage Oscilloscope and an isolation transformer to float the scope. The function generator creates a continuous sawtooth waveform. The frequency, output level and the ramp time can be adjusted as desired. The voltage ramp from the function generator goes to the input of a Kepco model BOP 500M bipolar amplifier (± 500 V, ± 80 mA max), which amplifies the signal by a factor of 100 and adds an adjustable DC bias to it. The driving voltage for is created by the function generator and amplified by the Kepco amplifier. The resultant sweeping voltage (usually set from about -40 volts to +40 volts) will drive the probe.

![Figure 3.10 Schematic of the Langmuir probe circuit used in this work.](image)

The oscilloscope has been intentionally isolated from the common/true ground by using an isolation transformer between the scope and the wall voltage. As seen in Figure 3.10, the scope
ground is instead connected to the output of the Kepco amplifier, say, as a “floating ground”. The outer conductor of the BNC connectors for all the channels will be at this ramped voltage. The coaxial cable from the probe is attached to channel 1 via a T-connection so that a 50 ohm terminator can be attached. This connects the center conductor (which goes to the probe) to the scope “floating ground” through a 50-ohm resistor, so that the channel 1 measures the voltage across the 50 ohm resistor and the probe current can thus be easily calculated. In fact, this current is relatively small that the probe nearly follows the sweeping scope voltage, differing by a few volts at most. Meanwhile, scope channel 4 measures the voltage of true ground, which is the opposite of the scope's floating voltage (sweeping voltage output). During a data run, both the probe current(s) and the sweeping voltage are measured by the scope. Often, multiple traces will be averaged in order to reduce random noises.

3.4.2 Triple Langmuir Probe

For the pulsed plasma diagnostics, triple Langmuir probe (TLP) was adopted to measure the electron density \( n_e \) and electron temperature \( T_e \). Unlike single Langmuir probe, TLP method does not require sweeping the voltage on probe tips and can be used for quick determination of temporal \( n_e, T_e \) \cite{79,80}. As illustrated in Figure 3.2, a fixed voltage \( V_{13} \) is applied between two of the probes (probe 1 and probe 3) and probe 2 is set to electrically float in the plasma. The current flowing between probe 1 and probe 3 is measured. Essentially, triple Langmuir probe only uses three points on a typical single Langmuir IV curve, as shown in Figure 3.11. \( V_2 \) is equal to \( V_f \). \( V_3 \) is deep in the ion saturation region to collect the ion saturation current, \( V_1 \) is slightly more positive than \( V_f \) to satisfy \( |I_1| = |I_3| \).
TLP is a well-studied technique, based on the assumptions of a Maxwellian electron energy distribution function (EEDF), collisionless thin sheath, and no interaction between probe tips [79,80]. The Maxwellian EEDF is a commonly used model of plasmas. More discussion concerning this assumption will be given later. The latter two assumptions could be satisfied by appropriate selection of probe tip diameter and the distance between tips [56]. Electron density and temperature can then be calculated using Equ. 3.1 and 3.2 [79,80]. Here $|I_{13}| = |I_1| = |I_3|$.

\[
\frac{1}{2} = \frac{1 - \exp\left(-\frac{eV_{12}}{kT_e}\right)}{1 - \exp\left(-\frac{eV_{13}}{kT_e}\right)} \tag{3.1}
\]

\[
I_{13} = \exp\left(-\frac{1}{2}\right) A e n_e \sqrt{\frac{kT_e}{M}} \tag{3.2}
\]

Triple Langmuir probe (TLP) are built as described in Refs. [56,61]. Three tungsten wires (0.25 mm in diameter) shielded by a multi-bore alumina tube are used as the probe tips. The exposed lengths of the probe tips are 8.8 mm and the separation of probes is 1.6 mm. Such dimensions are chosen to satisfy the collisionless thin sheath criterion and to prevent the interaction between probe tips. For a typical plasma condition (1 Pa, 10 eV and $5 \times 10^{17}$ m$^{-3}$), the sheath width (about 4 times Debye length) is estimated as 0.13 mm, smaller than the separation of tips, so that the interaction effects among the probes are negligible. The mean free path of electron-neutral collisions is calculated to be 8.10 mm, much larger than both the thickness of sheath width and the probe diameter to satisfy
the collisionless thin sheath criterion. In order to use the probe in a strong metal deposition environment, each probe tip is kept centered in the bore without contacting the metal coated alumina tube. This is also important for keeping the stray capacitance between probe tips and ground as low as possible [79].

Probe 1 and probe 3 are connected through a battery pack of about 58 V to meet one of the assumptions used to calculate the electron density and temperature, i.e., $V_{13}$ should be several times the electron temperature [80]. The current collected by probes 1 and 3 is obtained by measuring the voltage drop across a resistor in series (between 100 Ω and 10 kΩ depending on the plasma density). This voltage drop ($V_{34}$) and the voltage difference between probe 1 and probe 2 ($V_{12}$) are measured using differential probes (Tektronix P5200, with 50:1 attenuation). The signals are recorded by an Agilent Infiniium oscilloscope (1 GHz, 4 GSa/s). The oscilloscope ground is isolated from the wall outlet by an isolation transformer and then connected to the chamber ground, which greatly reduced the 60 Hz noise from the wall voltage.

The three probes are parallel to the target surface and are capable of moving in both axial ($z$) and radial ($r$) directions, as illustrated in Figure 3.2. In the present study, a region radially from $r = 0$ (the central axis) to $r = 14$ cm (near the chamber wall) and vertically between $z = 1$ cm (target at $z = 0$) and $z = 13$ cm (substrate at $z = 14$ cm) is scanned. The 3-D measurements are used to study the plasma distribution and transport.

It should be noted that, however, the triple Langmuir probe theory is based on the assumption of Maxwellian electron energy distribution function (EEDF). From Figure 3.11, it can be seen that the triple Langmuir probe only collect high energy portion of the electrons (between $V_f$ and $V_l$) and the electron temperature is calculated based on the EEDF in this energy region. A deviation from Maxwellian distribution, for example, a high electron energy tail can lead to an over-estimation of $T_e$ (or the effective electron temperature as a more accurate term to use here). Different research groups have reported that the pulsed discharge produces high-energy electrons initially, which quickly (within 10-20 μs) evolve into Druyvesteyn [81], bi-Maxwellian [40], or Maxwellian [82] distribution due to frequent ionization collisions and Coulomb collisions given a substantially high plasma density. There is a foreseeable error in the estimation of $T_e$ during the initial stage of discharge due to the large
high-energy electron population. This period of the discharge is not the main focus of the present study though. In the following part of the pulse and the off-time, the EEDF can be assumed to be approximately Maxwellian based on the fact that the determined $T_e$ is typically lower than 4 eV (as shown in the later section), indicating an effective relaxation of the hot electrons. In addition, the EEDF has been reported to be truncated at the energies of 4-7 eV [40, 81], so the over-estimation of the $T_e$ due to high electron energy tail should be small. With $n_e$ only weakly depending on $T_e$ ($n_e \propto T_e^{-1/2}$), the error of calculated $n_e$ induced by the error in $T_e$ determination is less significant. Nevertheless, the triple Langmuir probe data should always interpreted with caution.

3.4.3 Gridded Energy Analyzer/QCM Assembly

For the study of fluxes of various species in the plasma, a gridded energy analyzer (GEA) combined with a quartz crystal microbalance (QCM) [10, 83] is designed and placed in the downstream plasma. Its schematic is shown in Figure 3.12. The GEA is placed above the QCM and has a ceramic casing with an inner diameter of 30 mm. Three layers of stainless steel meshes are evenly placed inside with a gap of 6.4 mm. The wire distance in the mesh is 0.282 mm with 50% transparency. The distance between wires is chosen to be smaller than the sheath on the wire, which is estimated as 4 times the Debye length, to make sure the sheaths on the adjacent wires overlap to avoid the plasma leak [83].

The top mesh grid is usually floating to minimize disturbance to the plasma while the middle grid (the electron repeller grid) is negatively biased (typically at around -30 V) to reduce the electron penetration. The bottom grid (the ion repeller grid) is applied with an adjustable voltage from -50V to 30 V to admit or repel ions while the deposition rate of the total flux of metal atoms ($M^0$) and metal ions ($M^+$) or that of $M^0$ flux only is recorded by the QCM sensor. The QCM can also be biased negatively to make sure ions can reach the sensor. In order to isolate the QCM from ground, the metal water cooling tubes are replaced with plastic ones. The ground shielding of the QCM signal coaxial cable is cut off, while a capacitor of about 1 μF is added in between the cut-off ends to complete the path for high frequency QCM oscillation signal. If the QCM sensor is replaced with a current collector, the ion current of $Ar^+$ and $M^+$ can be measured. The flux of $M^0$, $M^+$, and $Ar^+$ can then be deconvoluted. Important parameters such as metal ionization fraction and ratio of $M^+$ flux to $Ar^+$ flux
can be determined. This setup may also be used to measure the energy distribution of ions.

The total fluxes of metal ions and neutrals $\Phi_{\text{tot,QCM}}$ admitted by the QCM sensor can be determined when the ion repeller grid is negatively biased. Then a positive bias is applied to the grid and gradually increased up to 30V so that only the neutral flux $\Phi_{N,QCM}$ reaches the QCM. The ion flux $\Phi_{\text{ion,QCM}}$ received by the QCM is obtained by subtracting the neutral flux from the total flux as given in Equ. 3.3. To calculate the actual ion flux $\Phi_{\text{ion,plasma}}$ and neutral flux $\Phi_{N,\text{plasma}}$ before entering the GEA, the transparency $T_g$ of the mesh grids is also taken into account as given in Equ. 3.4 and 3.5.

However, for the non-directional neutral flux, the ceramic casing will shadow part of the neutrals from reaching the QCM sensor, which induces a geometrical factor $G$. A more specific description of the calculation of $G$ was done by Green et al [83]. For the current GEA setup, $G=0.42$ is used in the calculation assuming an isotropic distribution of neutrals. For normally incident ions governed by electric field, the fluxes at the QCM sensor and that at the substrate are the same if there are no screens present and the factor $G$ is simply unity as implied in Equ.3.5. The $\Phi_{\text{ion,plasma}}$ and $\Phi_{N,\text{plasma}}$ are further used to determine the metal ionization fraction (IF) in the deposition flux as given in Equ. 3.6.

$$\Phi_{\text{ion,QCM}} = \Phi_{\text{tot,QCM}} - \Phi_{N,QCM} \quad 3.3$$

$$\Phi_{N,QCM} = \Phi_{N,\text{plasma}} (G) (T_g)^3 \quad 3.4$$

$$\Phi_{\text{ion,QCM}} = \Phi_{\text{ion,plasma}} (T_g)^3 \quad 3.5$$

$$I.F. = \frac{\Phi_{\text{ion,plasma}}}{\Phi_{\text{ion,plasma}} + \Phi_{N,\text{plasma}}} = \frac{\Phi_{\text{tot,QCM}} - \Phi_{N,QCM}}{\Phi_{\text{tot,QCM}} - \Phi_{N,QCM} + \Phi_{N,QCM} / G} \quad 3.6$$
3.4.4 Through-target Flux Diagnostics

Self-sputtering is believed to be a crucial feature of HiPIMS, as indicated by the OES measurements. Enhanced ionization of the sputtered metal neutrals leads to a great amount of metal ions returning with high energy and sputtering the target. This is especially a significant process for metals with high self-sputtering yield such as Cu. In this work, experiments will be designed to directly measure the fluxes of the incident species onto the target surface. The concept is to sample the plasma through a small orifice on the target. The tests will shed more light on the ionization process in the vicinity of target, in addition to the commonly used OES which is lack of direct quantitative characterization of fluxes and spatial resolution.

A special setup is designed with some magnetron modifications, as illustrated in Figure 3.13. A small orifice of 2.5 mm in diameter is drilled on the target. The incident plasma including Ar neutral and ions, metal neutral and ions and electrons can penetrate the orifice and reach the other side of the target. The vacuum of the system is maintained by enclosing the orifice with an additional small test chamber. A ceramic disk is placed between the stainless test chamber and the cooling plate for electrical insulation. Two sets of o-rings are used on both sides of the ceramic disk for vacuum sealing.

![Figure 3.13 Schematic diagram of the assembly for through-target fluxes measurement.](image)
For the flux measurements inside the test chamber, one layer of mesh is placed right after the orifice. It can be biased to as low as -600 V to repel the electrons (as the electron repeller grid). The ion flux penetrating through the mesh can be collected on an optional plate at about 10 mm away from the electron repeller grid. If the plate is removed, the ion flux can also be measured on the QCM. Here, the QCM has already been isolated from electrical ground and can be biased to as low as -600 V. Independent biases are used on the grid, plate and QCM. A ceramic tube is used to shield the grid, the plate from other biased surfaces such as the cooling plate at target voltage and the stainless steel test chamber at the QCM biasing voltage. Other than the current, QCM is also be used to measure the deposition rate. By biasing the QCM at proper voltages, the metal neutral and ion fluxes can be distinguished. As the fluxes travel toward the QCM, they can get scattered. To learn about this effect, Si wafers can be placed along the sidewall of the ceramic housing to monitor the deposition. The deposition thicknesses on Si are measured using cross-section scanning electron microscope (SEM).

The orifice location is right in the race track, as can be seen in Figure 3.14 (a). The choice of location was limited by the geometry of the magnet pack, since the magnet pack had to be cut to fit in the test chamber. Figure 3.14 (b) shows the test chamber. The electron repeller grid mesh, the ceramic shielding tube and the water-cooled QCM can be seen.

![Figure 3.14 Modification of the magnetron and installation of the test chamber. (a) Orifice was drilled on the Cu target, within the race track region. (b) The test chamber included grids shielded by ceramic parts and a water-cooled QCM.](image)
Chapter 4  HiPIMS Study on Planar Magnetron

In this chapter, fundamental studies of HiPIMS discharge are conducted in the Galaxy planar magnetron. Different aspects of the discharge are measured, including the IV characteristics, plasma parameters \((T_e, n_e, V_f)\), metal ion fractions, and deposition rates. Effects of various pulsing and discharge parameters are studied. Triple Langmuir probes are used to reveal temporal behaviors of the pulsed plasmas. By adding spatial measurements using a 3-D TLP, a picture of the plasma transport (expansion across the chamber) can be depicted. Not so surprisingly, both the pulsing parameters and the magnetic field configurations are found to affect the plasma evolution and transport. Ionization fractions in the deposition flux, as one of the most concerned properties of HiPIMS discharge, are determined using the gridded energy analyzer/QCM assembly. Finally the plasmas in the vicinity of the target are diagnosed via an orifice through the target. The returning fluxes towards the target are for the first time directly measured, providing new insights into the self-sputtering effect, etc.

4.1  HiPIMS Tests Using Al Target

Initial tests are performed in Galaxy planar magnetron system using an Al target. The goal is to check the functionality of the Huettinger plasma generator and the diagnostic tools. Different pulsing parameters such the charging voltage \(V_{ch}\), pulse duration \(t_p\), and repetition frequency \(f\), along with some discharge parameters such as pressure are varied. The magnetic field configuration used here is the horseshoe-shaped 300GL. Table 4.1 lists the completed tests in this section.

<table>
<thead>
<tr>
<th>Table 4.1 HiPIMS tests in Galaxy planar magnetron using Al target.</th>
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<td><strong>Pulsing parameters</strong></td>
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4.1.1  Discharge I-V Characteristics

Discharge I-V characteristics are measured to find its dependence on various parameters. The
waveforms of discharge voltage and current under different charging voltage and pulse duration are shown in Figure 4.1. First, the charging voltage $V_{\text{ch}}$ is increased from 500 to 1000 V, with the pulse duration fixed at 50 µs, and repetition frequency at 100 Hz. The Ar pressure is maintained at 5 mTorr. All the voltage waveforms show some oscillations right after the pulse beginning likely due to the capacitance in the circuitry. For $V_{\text{ch}}$ of 500 V, the discharge voltage is basically maintained at about 500 V, with a slight decrease. For a higher $V_{\text{ch}}$, the discharge voltage does not sustain at the set value. Instead, it quickly decreases to between 500 and 600 V within 15 µs. The discharge voltage is limited by the plasma itself with its impedance rapidly reduced during the discharge. Overall, a higher charging voltage still results in a higher discharge voltage on the cathode at any time during the pulse. And as a result, the pulse current is seen to scale with $V_{\text{ch}}$. A very high peak current of about 750 A is achieved with $V_{\text{ch}}$ at 1000 V. This is about two orders of magnitude higher than the typical DC discharge current. Within the pulse, the currents are seen to ramp up almost linearly versus time, showing no sign of saturation.

![Figure 4.1 HiPIMS discharge I-V characteristics vs. (a) charging voltage and (b) pulse duration.](image)

**Figure 4.1** HiPIMS discharge I-V characteristics vs. (a) charging voltage and (b) pulse duration. Discharge parameters are (a) 500-1000 V, 50 µs, 100 Hz, 5 mTorr, (b) 750V, 25-150 µs, 100 Hz, 5 mTorr.

Figure 4.1 (b) shows the results of different pulse durations between 25 and 150 µs. Other parameters are $V_{\text{ch}}$ at 750 V, f at 100 Hz, and pressure at 5 mTorr. The initial discharge voltage drop is also seen and then it stays at about 500 V. The discharge voltage waveforms of longer pulses overlap well with the shorter ones. The peak current keeps increasing during the pulse for those shorter than 75 µs. For longer pulses, however, the current saturates at about 500 A and starts to decrease after 75
It can be imagined that if the pulses are very long, the discharge will eventually evolve into DC mode with lower voltage and current on the cathode.

**Figure 4.2** HiPIMS discharge I-V characteristics vs. pulse repetition frequency. Discharge parameters are 750V, 50 µs, 25-300 Hz, 5 mTorr.

**Figure 4.3** HiPIMS discharge I-V characteristics vs. pressure. Pulsing parameters were 750V, 50 µs, 100 Hz, 1-10 mTorr.

The dependence of discharge IV characteristics on the repetition frequency is studied. The frequency is found to have little effect on the discharge voltage and current waveforms in a wide range (Figure 4.2). This is not difficult to understand since the pulse-off time is quite long for HiPIMS (10 milliseconds for typically used 100 Hz). The plasma generated by one pulse significantly diminishes over this off-time and thus has no effect on the next pulsing.

The pressure is varied from 1 to 10 mTorr using the same pulse recipe 750 V, 50 µs, 100 Hz.
At higher pressure, the discharge is maintained at lower voltage and higher current as shown in Figure 4.3. This is commonly observed even in DC discharge. The blowup in the first 10 µs also shows that the current is established earlier. At the beginning of the pulse, electron are emitted from the target and accelerated by sheath to have several hundred eV. A higher density of Ar atoms leads to more frequent electron collisions with the atoms, e.g. a higher ionization rate. These ions are then collected to form a more rapidly increasing current at higher pressure.

### 4.1.2 Temporal Behaviors of Pulsed Plasmas

In HiPIMS discharge, it is important to have time-resolved characterizations of the transient plasma. Triple Langmuir probe is adopted as a very convenient tool for this purpose. In this section, the initial tests of our house-made TLP will be described. The experiments are done in the Galaxy planar magnetron using Al target. Different recipes with varied charging voltage, pulse duration and working pressure are tested. The probes are kept at a same location, above the center of the substrate (r=0, z=13 cm).

In the first group of tests, the charging voltage is varied from 500 to 1000 V. The other conditions are 50 µs, 100 Hz, 5 mTorr. The measured signals of $V_{12}$ and $V_{34}$ are shown in Figure 4.4. According to Equ. 3.1 and 3.2, $V_{12}$ determines the electron temperature $T_e$ and $V_{34}$ determines the electron density $n_e$. It should be pointed out that when there is no plasma, $V_{12}$ is at about half the battery voltage. Calculation using this $V_{12}$ will give a very high electron temperature, though it is meaningless since there is no plasma in the first place. A similar situation will occur when the plasma is weak, i.e., $V_{12}$ tends to increase back to its initial value to cause an artificially high $T_e$. And this is the case observed in Figure 4.4 that $V_{12}$ signals shoot up after 50 µs. The high peak simply indicates a very weak plasma in the mean time. The $V_{34}$ signal, which is proportional to the current collected between probe 1 and 3, can be seen to increase during the pulse, decrease slightly after the pulse ends, and rise again to form a higher peak or peaks. More discussions will be given after the electron density calculation.
Figure 4.4 Examples of the $V_{12}$, $V_{34}$ signals for the triple Langmuir probe measurement. Discharge parameters are 600-1000 V, 50 µs, 100 Hz, 5 mTorr.

Figure 4.5 Evolution of $T_e$ and $n_e$ of HiPIMS plasma during pulses vs. charging voltage. Discharge parameters are 500-1000 V, 50 µs, 100 Hz, 5 mTorr.

Figure 4.5 shows the calculated $T_e$ and $n_e$ during the 50 µs pulse and their dependences on the charging voltage from 600 to 1000 V. Again, the $T_e$ and $n_e$ estimation is based on some assumptions. The possible error by TLP measurements has been discussed in Section 3.4.2. The $T_e$ data in the first 10 to 15 µs should not be used since the growing plasma is far away from Maxwellian distribution in this period. After 15 µs, $T_e$ shows a generally decreasing trend. Under a higher charging voltage, $T_e$ decreases to a lower value at the pulse end. For example, $T_e$ drops to about 4 eV in the 600 V recipe and to about 2.5 eV in the 1000 V recipe. A surge of $n_e$ is observed from the initial time indicating the establishment of the plasma. The spike might be caused by very high electron energies of several
hundred eV even though the electron density is actually low. Later on, \( n_e \) gradually increases. The higher is the charging voltage, the more quickly \( n_e \) ramps up. Up to \( 5 \times 10^{17} \text{ m}^{-3} \) is measured using the 50 \( \mu \text{s} \) pulses. The opposite trends for \( n_e \) and \( T_e \) have been commonly observed in plasma. Increased plasma density leads to more frequent Coulomb collisions for the electrons to dissipate energies.

Figure 4.6 Temporal behaviors of \( T_e \) and \( n_e \) using different charging voltage during and after the pulse. Pulse duration is 50 \( \mu \text{s} \).

Figure 4.6 shows the calculated \( T_e \) and \( n_e \) later into the pulse-off time. Compared with Figure 4.4, \( T_e \) and \( n_e \) do have similar shapes as \( V_{12} \) and \( V_{34} \) signals respectively. \( T_e \) keeps decreasing to very low values. This is because the late-arrival electrons go through a lot of collisions while diffusing toward the substrate, and keeps losing energy during the time. The \( T_e \) peaks between 50 \( \mu \text{s} \) and about 150 \( \mu \text{s} \) are artificial because of the reason explained above. The plasma is too weak in the meanwhile so that \( V_{12} \) tended to return to the value of about a half of \( V_{13} \) as in the case of no plasma present. This might cause an under-estimation of \( n_e \) by a factor of 2 to 4. But it should not change the overall \( n_e \) behavior that it drops after pulse ends at 50 \( \mu \text{s} \) and soon goes up again to form a high second peak. It is believed that this peak is from the plasma expansion out of the magnetic confinement region. The magnitude and the arrival time of this peak vary with the used recipes. The expansion peak \( n_e \) scales with higher \( V_{ch} \). Higher than \( 2 \times 10^{18} \text{ m}^{-3} \) is achieved for 1000 V recipe. This is easy to explain since higher-\( V_{ch} \) recipes generate denser plasmas in the race track during the pulse as the source of expansion. The peak arrives sooner for higher-\( V_{ch} \) pulses, i.e. the expansion is faster. This might be caused by more Coulomb collisions and consequently higher diffusion constant across the magnetic field \( D_{\perp} \) towards the substrate. More discussions on this will be given in Section 4.2.2.
there are actually two peaks very close to each other. The later one is believed to be from expansion plasma diffusing backwards after reaching the substrate. More discussions are given in Section 4.2.1.

Similar measurements were performed for recipes with different pulse duration. The $T_e$ and $n_e$ results are shown in Figure 4.7. During the pulse, $T_e$ follows the same decreasing trend and overlaps very well with each. And $n_e$ started to gradually increase after 25 $\mu$s. The longer the pulse, the higher $n_e$ was achieved, for example, about $1.5 \times 10^{19}$ m$^{-3}$ for 150 $\mu$s at the end of pulse. Increasing the pulse duration seems to be an effective way to enhance the plasma density.

After the pulse, there are the similar artificial $T_e$ signals, especially for the shorter-pulse recipes. The shorter is the pulse duration, the longer the plasma is absent. This is consistent with the observance that second plasma peak in shorter-pulse recipes takes longer to reach to the substrate. For pulses longer than 75 $\mu$s, however, the values of peak arrival time are about the same. It is suspected that the plasma starts to effectively diffuse out of the race track region when the density in the race track is high enough (so the cross-B diffusion constant $D_\perp$ is high enough) and not necessarily when the pulse ends. Based on Figure 4.1 (b), the discharge current reaches maximum at about 75 $\mu$s, which means the electron density in the race track peaks at the same time and starts to diffuse out at about the same time for all the recipes of 75 to 150 $\mu$s pulses. This is consistent with the fact that there is no artificial $T_e$ peak for a longer-pulse recipe since plasma has started expansion even before the pulse ends.

![Figure 4.7 Temporal behaviors of $T_e$ and $n_e$ using different pulse durations. Discharge parameters were 750 V, 25-150 $\mu$s, 100 Hz, 5 mTorr.](image)

The effect of pressure on $T_e$ and $n_e$ was measured by using the same pulse recipe 750 V, 50 $\mu$s,
100 Hz. As shown in Figure 4.8, $T_e$ also obviously decreases during the pulse. The values of $T_e$ and $n_e$ at different pressures during the pulse are comparable considering the error bars. In fact, they have similar discharge current. 10 mTorr discharge has a longer plasma absence during the pulse-off time, which is consistent with the longer delay time for the expansion peak to arrive. This is likely due to more pronounced scattering at higher pressure to reduce the diffusion constant.

![Figure 4.8 Temporal behaviors of $T_e$ and $n_e$ at different pressure. Discharge parameters were 750 V, 50 $\mu$s, 100 Hz, 1-10 mTorr.](image)

### 4.1.3 Spatial Distributions of Pulsed Plasmas

The above TLP measurements show that the pulsed plasma does not disappear immediately after a pulse ends, but takes some time to disperse to the substrate (and other walls) to form a second peak. To better depict this expansion process, it is helpful to measure the plasmas at different distances from the target. Here, three locations of $z = 3.5, 8.5, 13$ cm along the center axis of the chamber were used.

The comparison is first made using a set of recipes with different charging voltages, as in Figure 4.9. Some obvious changes can be seen as the TLP moves from $z = 3.5$ cm (closer to the target) to $z = 13$ cm (closer to the substrate). First, the electron temperatures are higher near the target. This is probably because of the high energy electrons escaping the confinement which haven’t had many collisions to lose energy yet. Second, the densities during the pulse are higher near the target, which is expected. Third, at $z = 3.5$ cm, the second peak $n_e$ are about twice higher than $n_e$ at the end of the pulse. For some recipes like 900 V, there is no drop of $n_e$ at the pulse end. This probably suggests that the diffusion starts earlier in the pulse. At $z = 13$ cm, the second peak $n_e$ are usually 20 times higher.
than \( n_e \) during the pulse.

Figure 4.9 Triple probe measurements at different locations (3.5, 8.5, 13cm from the target). Discharge parameters were 600-900 V, 50 µs, 100 Hz, 5 mTorr.

It is believed that electrons reaching \( z = 13 \) cm are those high energy electrons, which have a much smaller population than the confined electrons diffusing out later. Fourth, the second \( n_e \) peaks appear earlier at \( z = 3.5 \) cm and much later at \( z = 13 \) cm. The peaks also become broader, indicating a larger number of collisions encountered. Finally, it can be seen that the second \( n_e \) peaks appear at
about the same time at $z = 3.5$ cm for all the recipes, but much later for the 600 V recipes. In another word, the plasma expansion may have similar speeds near the target but is slowed down more for the lower-$V_{ch}$ recipes.

Figure 4.10 Triple probe measurements at different locations (3.5, 8.5, 13 cm from the target). Discharge parameters were 750 V, 50-150 $\mu$s, 100 Hz, 5 mTorr.

For different pulse durations of 50-150 $\mu$s, TLP was also performed at the three locations, as
shown in Figure 4.10. Similar analyses can be done as above to clearly show the plasma expansion process. One interesting point is that the 150 μs pulse shows no second peak at z = 3.5 cm. Instead $n_e$ maximum is at about 100 μs during the pulse. This further supports the previous statement that the plasma expansion starts when the density is high enough, not necessarily at the pulse end.

### 4.1.4 Deposition Flux Measurements

Ionization fraction measurement was tested using the GEA/QCM assembly. The QCM was placed on the substrate. A recipe of 750 V, 100 μs, 100Hz and 5mTorr was used. Top grid was kept floating and the electron repelling grid was at -20 V (lower than the floating potential $V_f$). The ion repelling grid was biased between -30 V and +30 V. The metal deposition rates under different ion repeller grid biases were measured as shown in Figure 4.11. It can be seen that the deposition rates level out when the bias is lower than -20 V or higher than +20 V, corresponding to the total flux of Al atoms and ions, and the flux of Al atoms only. Using these two deposition rates, Al ionization fraction in the deposition flux can be calculated using Equ. 3.6 to be about 29 ± 4 %. This fraction is quite high as compared with the DC magnetron sputtering in which the ionization fraction is usually just several percent. More results on ionization fractions (of Cu) will be given later in Section 4.3.

![Figure 4.11](image)  
**Figure 4.11** Ionization fraction measurements using the GEA/QCM assembly on the substrate level. The discharge parameters were 750 V pulse voltage, 100 μs pulse length, 100Hz and 5mTorr Ar.

The deposition rates by HiPIMS were measured using QCM under different conditions of varied charging voltage, pulse duration (or length), repetition frequency and pressure. For the purpose of comparison, they are plotted in Figure 4.12 against the average power. The deposition rates in different categories basically track with the power, except in the varying pressure case. At 2 kW and 5
mTorr, the deposition rate is about 25 nm/min. These deposition rates will be compared with those in MPP sputtering.

Figure 4.12 The deposition rates vs. average power in HiPIMS, tested under different pulse voltage, pulse length, repetition frequency, and pressure.
4.2 Plasma Transport Study

In the initial tests of TLP measurements, the plasma expansion was clearly observed with a second $n_e$ peak moving away from the target. High-density plasma diffuses across the magnetic field toward the substrate. However, without a detailed spatial characterization of the plasma, it is difficult to predict the plasma transport in the 3D space, for example, the orientation and the speed at different positions. The non-uniform magnetic field configuration only complicates the task. On the other hand, it is crucial to further understand the plasma transport mechanism for films deposition. It determines the fluxes of plasma species including ions received on the substrate level, the time delay of the fluxes and their uniformities. 3-D characterizations of pulsed plasmas using triple Langmuir probe are thus performed. The influences of pulsing parameters and equally important magnetic field configurations are studied.

4.2.1 3D Plasma Characterization

First, the HiPIMS discharge in Al target using the 300GL magnetic field configuration is still used to demonstrate the 3D TLP measurements. The triple probes were scanned in the chamber radially from $r = 0$ (the central axis) to $r = 14$ cm (near the chamber wall) and vertically between $z = 1$ cm (near the target) and $z = 13$ cm (near the substrate). The radial axis is chosen to be opposite to the opening point in horseshoe-shaped race track. An example of moving TLP radially is shown in Figure 4.13. The probes are at kept at $z = 13$ cm, or 1 cm above the substrate. The recipe used is 800 V, 50 µs, 100Hz and 5mTorr. Only temporal $n_e$ curves are shown here. Similar structures can be seen including the $n_e$ ramp-up in the pulse and a second $n_e$ peak afterwards. To quantify the measurements, the $n_e$ at the end of the pulse, second $n_e$ peak value, and the peak delay time after the pulse end were determined. Figure 4.14 shows another example of moving TLP from the target vicinity towards the substrate along the central axis ($r = 0$). As expected, the expansion peak appears later and later while its amplitude becomes lower. One interesting observation is the multiple peaks seen in the expansion plasma. As marked in the figure, there appears only one peak at $z = 13$ cm, while two peaks show up at $z = 11$ cm. They become further apart when measured at $z = 8$ cm. It is believed after the expanding plasma reach the substrate, the substantially increased density there results in the plasma diffusing
which is directed back. This is evidenced by the later appearance of the second peak in the pair when the probes are farther from the substrate. Similar double peak structures have been seen in Figure 4.6. In this case, the probe is about 1.2 cm higher than substrate, so the peaks are very close.

![Figure 4.13](image1.png)

**Figure 4.13** Triple probe measurements at different r positions at z = 13 cm (1 cm above the substrate). Discharge parameters are 800 V, 50 µs, 100 Hz, and 5 mTorr.

![Figure 4.14](image2.png)

**Figure 4.14** Triple probe measurements at different z positions along the center axis (r=0). Discharge was performed in planar magnetron using the 300GL configuration and recipe of 750 V, 50-150 µs, 100 Hz, 5 mTorr.

With the data measured at many different points in the chamber, a mapping of the pulsed
plasma density can be done at any temporal location. Since we are more interested in the plasma transport, the plasma from the end of the pulse ($t = 50 \mu s$) to as late as $t = 190 \mu s$ are mapped and compared, as in Figure 4.15. It should be noted that the scale of color map is different from each other based on the maximum $n_e$ at the time being, so the overall densities in $t = 190 \mu s$ map are much lower.

Figure 4.15 Plasma ($n_e$) mapping in the r-z plane and its evolution after the pulse end.
The comparison provides a visual image of the plasma expansion over time. It starts with two spots of high density plasmas, corresponding to the two race track circles. The outer race track (at $r = 11$ cm) has denser plasmas at the end of the pulse as compared with the inner one (at $r = 4$ cm). The expansion at the beginning seems to be straight downwards but gradually become more isotropic. The expansion from the outer race track seems to move faster, reaching the substrate at about $t = 110 \mu s$. The inner one moves relatively more slowly, leaving a “hot” zone in the lower central region even at $t = 190 \mu s$.

The density mapping at different times is helpful for visualization but is cumbersome and not quantitative. So instead the $n_e$ at the end of the pulse, second $n_e$ peak value, and the peak delay time after the pulse end were extracted at different locations and plotted together for comparison. Figure 4.16 (a) and (b) shows the $n_e$ at the end of the pulse and expansion peak $n_e$ value. The two high $n_e$ peaks at $t = 50 \mu s$ are $2.6 \times 10^{19}$ m$^{-3}$ and $2.6 \times 10^{19}$ m$^{-3}$, reflecting the race track positions. The expansion peak $n_e$ values for these two positions are $2.8 \times 10^{19}$ m$^{-3}$ and $5.7 \times 10^{19}$ m$^{-3}$. This very dense plasma is expected to produce highly-ionized metal flux and induce the self-sputtering effect. The two $n_e$ values near the target are about the same, meaning $n_e$ almost immediately starts to drop after the pulse ends. This is reasonable since the density near the race track region can only decrease after the discharge with all the plasma diffusing out. The location between the two race tracks near the target has a very low density even after expansion, implying it was very difficult for the plasma to diffuse laterally. As plasma expanding from the target towards the substrate, the peak $n_e$ rapidly decreased while the distribution in a same plane became more leveled. At $z = 11$ and $z = 13$ cm, the expansion peak densities are quite uniform, which is a great improvement as compared with those at $t = 50 \mu s$.

Figure 4.17 displays the peak delay time at varied locations. The expansion peaks at $r = 5$ cm, $11$ cm, and $z = 1$ cm appears only a few $\mu s$ after the pulse ended, meaning that the density starts to decrease immediately. The farther away from the race track, the longer it takes for the expansion peak to show up. The plasma expansion peaks also seem to move faster along the axis of $r = 10$ cm than along the center line ($r = 0$). About $150 \mu s$ is needed for the expansion plasma peak to reach the substrate. It should be noted that after the plasma density peak appeared, the plasma remained for a long period before completely fading away. It is thus important to have a long enough off-time between pulses to ensure the plasma including ion species to reach the substrate.
Figure 4.16 The spatial distribution of (a) $n_e$ at the end of the pulse ($t = 50$) and (b) expansion peak $n_e$ values. Recipe is 800 V, 50 µs, 100 Hz, 5 mTorr.

Figure 4.17 The peak delay time after the pulse at various locations. Recipe is 800 V, 50 µs, 100 Hz, 5 mTorr.

4.2.2 Effect of Magnetic Field Strength

The above example of the 3-D scanning triple Langmuir probe measurements shows that the expansion in the chamber depends on the race track shape and thus the magnetic field. In this section, experiments are performed to study three relatively simple configurations, 200G, 500G, and 800G
ring-shaped configurations, regarding the effect of B field strength on the plasma transport mechanisms. A Cu target is used instead of Al. Using the same method described above, the information of the expanding plasma such as the peak density and peak delay time at various positions was extracted. The data from 500G case are plotted in Figure 4.18. It shows the expansion from the only one race track with the \(n_e\) decreasing from a very high value of \(5.5 \times 10^{19} \text{ m}^{-3}\) to about \(1 \times 10^{18} \text{ m}^{-3}\).

![Figure 4.18](image)

Figure 4.18 The spatial distributions of (a) expansion peak \(n_e\) and (b) the peak delay time. Recipe is 800 V, 50 \(\mu\)s, 100 Hz, 5 mTorr. 500G configuration and Cu target are used.

![Figure 4.19](image)

Figure 4.19 Peak \(n_e\) and the corresponding peak delay time vs. various magnetic field strength (200-800 G), (top row) peak density of the expanding plasma, (bottom row) delay time of the expansion peak.

In order to better visualize the distributions of these two parameters, contour plots in R-Z plane are used based on the same data. Figure 4.19 shows the mapping of peak \(n_e\) and peak delay time.
of the expansion plasma in the 200G, 500G, 800G configurations respectively. In all three cases, peak $n_e$ near the target reflected the ring shape of the race track. The higher the magnetic field strength, the denser the plasma. A closer look at the figures revealed that as the B field strength increased from 200 to 800 Gauss, the expansion changed from being nearly isotropic to being more directional toward the substrate. In fact, as a result of the more directional expansion in the 800G configuration, the peak densities around the center axis of the chamber were overall very low, even lower than those in the 500G configuration. From the figures of the peak delay time, it can be seen a stronger B field led to faster expansion. Re-plotting the figures using different scales can provide more information as shown in Figure 4.20. Obviously, the 200G configuration leads to more isotropic distribution of the peak $n_e$ of the expanding plasma. It is also easier for a lateral diffusion as compared with the 800G case and has more comparable delay time when approaching the substrate.

![Figures showing peak density and delay time](image)

**Figure 4.20** Re-plotted peak $n_e$ and the corresponding peak delay time vs. various magnetic field strength.
In a magnetron plasma, electrons are confined by magnetic field via the $\mathbf{E} \times \mathbf{B}$ drift and the diamagnetic drift \cite{84}. The corresponding drift velocities, $\mathbf{u}_E$ and $\mathbf{u}_D$, are perpendicular to the field and the density gradients. Despite the magnetic confinement, electrons still move across the magnetic field due to drifting in the presence of electric field and diffusion. Both contribute to $\mathbf{u}_\perp$, the flux velocity perpendicular to $B_0$, as the first and second terms in the RHS of Equ. 4.1. $\mu_\perp$ and $D_\perp$ are the mobility and diffusion constants perpendicular to magnetic field, $n$ is the density, and $\mathbf{E}$ is the electric field. The last term of Equ. 4.1 describes the $\mathbf{E} \times \mathbf{B}$ drift and the diamagnetic drift. The diffusion is strongly retarded by the magnetic field. As can be seen in Equ. 4.2, $D_\perp$ is smaller than the diffusion coefficient without a magnetic field, $D$, by a factor of $1 + (\omega_c \tau_m)^2$. Here $\omega_c$ is the gyration frequency and $\tau_m \equiv 1/\nu_m$ ($\nu_m$ is the momentum transfer frequency), $\bar{r}_c$ is the mean gyroradius which is inversely proportional to $B$. A higher magnetic field thus has a lower diffusion coefficient to build up a higher-density plasma during the discharge.

\[
\mathbf{u}_\perp = \pm \mu_\perp \mathbf{E} - \frac{D_\perp \nabla n}{n} + \frac{\mathbf{u}_E + \mathbf{u}_D}{1 + (\omega_c \tau_m)^{-2}} \quad 4.1
\]

\[
D_\perp = \frac{D}{1 + (\omega_c \tau_m)^2} = \frac{\pi}{8} \bar{r}_c^2 \nu_m \quad 4.2
\]

Based on these two equations, it can be seen that two factors may play important role in determining the plasma flux velocity. One is the $D_\perp$ and the other is the electric field $\mathbf{E}$. For $D_\perp$, it also varies with the electron density due to the Coulomb collisions. At low density regime, this is negligible comparing to the electron-neutral collisions. However, when the plasma is very dense in HiPIMS situation (with $5 \times 10^{19}$ m$^{-3}$ measured 1 cm away from the target), this Coulomb collision leads to a greatly increased $\nu_m$, which reduces $D_\perp$. As a result, the diffusion across the B field becomes more effective. The non-classical diffusion has been observed in HIPIMS that the effective collision time for electrons is much shorter and cross-B diffusion is enhanced \cite{71,85}. So for 800G configuration, electron density can quickly build up but when it is high enough, it has a self-limiting mechanism due to the relationship between diffusion loss rate and the density. Same rule can be applied to recipes of high $V_{ch}$ or longer pulses. The high density leads to enhanced diffusion and contributes to a faster expansion.
Figure 4.21 The evolution of plasma floating potential at varied z positions (r = 10 cm) in (a) 200G, (b) 500G, and (c) 800G configurations.

For the drifting in the electric field, the potential distribution inside the chamber should be known. The floating potentials $V_f$ were therefore measured in all three configurations. Figure 4.21 shows the $V_f$ from $z= 1$ cm to $z = 13$ cm below the race track (R = 10 cm). Unlike in an equilibrium DC plasma where potential difference in the presheath and in the bulk plasma is small, a very large potential drop existed along the z direction at the beginning of the pulse ($t = 10$ µs). As time evolved, this potential drop gradually decreased and eventually a nearly flat potential was established between $z= 1$ cm to $z = 13$ cm. The time needed for this to occur was seen to depend on the magnetic field strength, about 40 µs in the 200G configuration, about 60 µs in the 500G, and more than 150 µs in the 800G configuration. At the pulse end, the potential differences between $z= 1$ cm and $z = 13$ cm were -2, -6, and -22 V. The higher electric field in the 800G configuration then prompted the drifting and increased the expansion speed.

In order to explain the preferred orientations, the spatial distributions of the $V_f$ were measured at $t = 40$ µs and 50 µs (Figure 4.22). At 800G, it can be seen that the floating potential is much lower near the race track region while much higher in the lower part of the chamber. This gradient leads to preferred direction for the electron drifting. While in the 200G case, the center part of the graph becomes lower (red). The electron diffusing out the race track won’t see a dragging force or even on the contrary some resistance to lead to slower and more isotropic expansion.

The magnetic field itself is another factor to likely affect the expansion direction. For the plasma to move out of the race track region, it had to diffuse across the magnetic field, either laterally in radial direction or in axial direction. As shown in Figure 4.23, magnetic field mapping showed that
$B_y$ rapidly decreases when moving away from the target, while moving in radial direction the magnetic field becomes stronger because of being closer to the magnets. So in the 800G field, it was relatively easier for plasma to diffuse axially, while almost prohibited to move in radial direction because of the even higher B field. This is a less significant factor for the 200 G configuration, since the overall weak B field still allowed the plasma diffusion in all directions.

Figure 4.22 The measured floating potential distributions in the chamber in (left) 200G, (middle) 500G, and (right) 800G configurations.

Figure 4.23 The magnetic field profile ($B_y$) between the target plane and substrate for (left) 800G and (right) 200G configurations.
4.3 Metal Ions in Deposition Flux

Plasma diagnostics have shown that the HiPIMS discharge generates very high density plasmas in the vicinity of race tracks. High ionization fractions are thus expected in these regions. Part of the ions will be attracted back to the target for the self-sputtering. Then the question is whether ions can be efficiently extracted towards the substrate for deposition. Undoubtedly, the metal ionization fraction in the deposition flux is a very crucial property for a HiPIMS deposition process. In this section, the metal ionization fractions are measured using the gridded energy analyzer/QCM assembly. Its dependence on various pulsing parameters and the magnetic field strength will be studied.

4.3.1 Cu Ionization Fractions

Using the same setup of GEA/QCM assembly, ionization fractions of Cu in the deposition flux were measured at the center of the substrate. The experiments are performed in 200, 500 and 800G configurations to study the effect of B field strength. The pulse charging voltage V_{ch} and pulse duration t_p are varied.

![Figure 4.24 Cu ionization fractions at the center of the substrate at different (a) charging voltage and (b) pulse duration. 200, 500, 800G configurations are used.](image)

The data are shown in Figure 4.24. It showed that Cu⁺ fraction increased with V_{ch} in all three B field designs, which was attributed to higher n_e. 200G configuration generated the highest Cu⁺ fraction in the deposition flux, up to about 60%. For most commonly used 500G magnetron, this fraction was reduced to about 40%. These seemed to contradict with the fact that stronger magnetic
field produced much denser plasma. However we found this may be caused by the different plasma potential distributions in these magnetic fields. The effect of \( t_p \) was complicated and depended on the B field. At 200G, Cu\(^+\) fraction basically decreased with longer pulses. At higher B field, it seemed to increase accordingly.

### 4.3.2 Discussions

In a DC magnetron sputtering plasma, a relatively short presheath of about 1cm forms in front the racetrack, in which ions are generated. Beyond this presheath region, the plasma potential is relatively flat so ions can diffuse out to the substrate. However, this is the case in HIPIMS due to the transient nature of the pulsed discharge. The floating potential \( V_f \) measured at varied positions and time are shown in Figure 4.25. Generally \( V_f \) should track with the plasma potential \( V_p \) giving the electron temperature doesn’t vary too much. In 200G configuration, it is seen that at the beginning of the discharge, a very large potential difference of about 40V exists between the target and the substrate. As a result, all the ions generated by the intense plasma in the racetrack will be directed towards the target and then get collected. These ions help the fast increase of discharge current but do not contribute to ion flux to the substrate. A flat potential is only established after 40\( \mu \)s between the target and the substrate. In another word, ions can get to substrate after 40\( \mu \)s. In 500G case, a larger potential gradient is seen and the flat potential is established later, after 50 \( \mu \)s. At 800G, flat potential exists only after 150 \( \mu \)s. And before that, only positions very close to the substrate may contribute to the ion extraction. All the points (of different positions and time) that contribute to the ion extraction are connected using bold lines. In another word, the effective ion extraction in a high B configuration only happens in a smaller region and a shorter period, resulting in a lower Cu ionization fraction in the deposition flux.
Figure 4.25 Floating potential measured at various locations between the target and the substrate, at different time from the start of pulse. 200, 500 and 800 Gauss configurations were compared. Bold lines show at what positions and at what time ions generated can be extracted to the substrate.

Similar measurements are performed for pulses of different lengths. Figure 4.26 shows at 200G a uniform plasma potential across the discharge region is established after 40 µs (lines at different “z” lie on top of one another). At 500G, it is from about 75 µs. At 800G, only between the substrate and a certain point lower than z=3 cm there is a flat potential (not shown here). Again, these determine that only ions generated at the pulse beginning and near the target will not diffuse to the substrate.

Figure 4.26 Temporal evolution of floating potentials measured at various locations (z=13cm is close to the substrate and z=3cm close to the target). 200, 500 and 800 Gauss configurations are compared.

The ionization rate during the discharge is then calculated based on the measured T_e and n_e data. HIPIMS has been claimed to have very high metal ionization due to its high density n_e (~10^{20} m^{-3}) during the pulse. Other than n_e, the electron temperature also matters since it changes the ionization rate K_{iz} for electron impact ionization.

\[
\frac{dn_M}{dt} = n_e K_{iz} n_M
\]

Here n_M is the metal atom density. Other ionization processes such as Penning ionization and charge exchange expansions (4.3)
exchange with Ar ions are less significant here. Figure 4.27 shows the calculated ionization rate $K_{iz}$ as a function of $T_e$ under a Maxwellian distribution of electron energy. The calculation is based on the cross-section data taken from [86].

![Figure 4.27](image)

**Figure 4.27** Cu ionization rate by electron impact as a function of electron temperature.

Based on the 3-D $T_e$ and $n_e$ measurements, $n_e K_{iz}$ were calculated. A more explicit parameter is the probability $P$ of an atom being ionized per unit distance (here as 1cm). Here $v_M$ is the average speed of metal atoms.

$$P = 1 - \exp(n_e K_{iz} / v_M)$$

This probability is calculated for varied positions in the plasma and at varied time, in all three B field strengths, as shown in Figure 4.28. There are several features in the plots.

1) The probability of ionization is very high, nearly 100%, at all positions in the chamber at the beginning of a pulse. This is due to the high-energy electrons which are not yet thermalized, even though the electron density is low. And the ionization probability quickly decreases with time, even in the racetrack region with an increased $n_e$. This is because of a quickly decreasing $T_e$ to about 1 or 2 eV.

2) The ionization probability closely depends on the positions. And the highest value is not necessarily in the racetrack region. In fact, the ionization occurs in the entire region and as time evolves $P$ becomes higher near the substrate than near the racetrack.

3) The 800G does not necessarily have higher ionization probability than 200G. Instead, it depends on the position and time. For example, the ionization probability near the substrate is higher in 200G, which is likely because high energy electrons escape more easily from the magnetic field confinement to reach the substrate region.

It is learned from Figure 4.25 that only ionizations at certain positions and from certain time
can contribute to the downstream ion flux. We herein mark those points in Figure 4.28 of probability of ionization. Only points connected by bold lines should be considered. All the other points lead to a returning ion flux to the target. It is then clear that with similar ionization probabilities, 200G has the largest region and longest period to extract ions. On the other hand, 800G can only extract ions after the pulse is already off and only from a limited region close to the substrate. It is thus not difficult to see why the meal ionization fraction measured on the substrate was the highest in 200G.

![Graphs showing probability of ionization per cm for 200G, 500G, and 800G.](image)

**Figure 4.28** Probability of atoms being ionized per cm. Calculations were done for different positions, time, and B field strengths. $z=14$ cm corresponds to the substrate level and $z=0$ is the target level.

The probabilities for Cu atoms being ionized per cm are also calculated for different pulse lengths. Figure 4.29 gives the probabilities at varied positions in the plasma and at varied time. Generally, ionization probability is lower for a longer pulse. But for higher B field configuration, especially for 800G, the ionization starts to increase again from 75 or 100 µs. This is predictable with a nearly constant $T_e$ but significantly increase of $n_e$.

In Figure 4.26, it is showed that 200G starts the ion extraction from 40 µs, so all the points from 40 µs on till the end of the pulse should be considered. A long pulse (e.g. 150 µs) will include all the points with very low ionization probability, and thus an overall lower ionization fraction as
compared with shorter pulses. However for higher B fields, for example 800G, the obvious increase of the ionization probability from 75µs leads to a higher ionization fraction with longer pulses. In short, the opposite trend of ionization fraction as a function of pulse length between 200G and stronger-B field configurations is due to the opposite trend of ionization rate.

![Figure 4.29 Probability of atoms being ionized per cm using 150µs pulses. Calculations are done for different positions, time, and B field strengths.](image)
4.4 Through-target Flux Measurement

Studies have shown HiPIMS has many distinctive features such as very dense plasma, high ionization degree of the sputtered materials, self-sputtering, rarefaction effect, etc. All these processes occur in the vicinity of the target, which makes it quite challenging to directly diagnose the plasma without significant disturbance. Indirect diagnosis methods such as optical spectroscopy or fast camera usually lack of good spatial resolution or quantitative characterizations. In this section, a new concept as to sample the plasma fluxes through an orifice in the race track region is tested.

The designs of the test chamber and all the modifications to the magnetron were described in Section 3.4.4. The orifice is located in a race track where the $B_\parallel$ is about 300 Gauss, as shown in Figure 4.30. Although it’s not in the region of the strongest B field, it turns out be advantageous for some tests.

![Figure 4.30 Location of the orifice as shown in the $B_\parallel$ mapping on the target surface.](image)

4.4.1 Ion Flux Measurement

The plasma through the orifice includes different species such Ar$^+$ ions, Cu$^+$ ions, electrons and neutral atoms. The ions are especially interested here. The grid and/or the optional plate are thus arranged in a setup similar to the previous described gridded energy analyzer (GEA). The first grid
after the orifice inlet is used as the electron repeller grid. The bias on it can be varied from 0 to -600 V. At about -500 or -600 V, all the electrons are repelled and only ions reach the grid, get collected or penetrate through the mesh. A current collecting Cu plate is installed after the first grid. It can be biased between +100 to -600 V. If biased negatively at about -500 V, all the penetrating ions are collected. The plate can be installed at different distances from the first grid. The QCM can also be biased and used to collect current, in which case, there is no need to install the optional plate.

Using a recipe of 900 V, 50 µs, 100 Hz, 5 mTorr, the currents are measured using this setup. First of all, the bias on the electron repeller grid is changed from -50 V to -600 V. The QCM serving as a current collecting plate is biased at -500 V. No Cu plate is installed. The currents on the grid are shown in Figure 4.31 (a). When the grid bias is -50V, the current is always negative during the pulse. Obviously the electrons are not completely repelled by the grid. As the bias decreases, the collected current becomes more positive. It eventually saturated at about -600 V when only ions are collected.

![Figure 4.31](image-url)  
**Figure 4.31** Current measured on the first grid at different bias on this grid using three different recipes. (a) 900 V, 50µs, 100 Hz, 5 mTorr. (b) 800 V, 50µs, 100 Hz, 5 mTorr, (c) 750 V, 50µs, 100 Hz, 5 mTorr
Similar tests are also performed for 800 V and 750 V pulses, as shown in Figure 4.31 (b) and (c). Comparison of the three recipes reveals that the ion current increases with higher charging voltage. This is easy to understand since the 900 V recipe produces much higher $n_e$ as measured in the previous section and consequently a higher discharge current. The electrons are completely repelled with a bias of -600 V in 900 V recipe, while in the 800 V and 750 V, the biases required for this are -500 V and -400 V. This is likely because the electrons are the secondary emission electron from the target. A bias lower than the target voltage is thus needed to repel these electrons. The discharge voltage on the target using 900 V recipe is indeed lower than the other two.

It is also noticed that the ion currents abruptly increase at a certain time during the pulse. As a comparison, the discharge currents on the target always smoothly increase. This time delay to observe
ion current also depends on recipes. It is about 31 \( \mu \text{s} \) for the 750V recipe, 21 \( \mu \text{s} \) for the 800V recipe and 14 \( \mu \text{s} \) for the 900V recipe.

The stronger recipe may lead to a faster drifting speed for the strong plasma zone. Or it enables the strong plasma to be established at a position closer to the hole where the B field there is not the strongest.

This abnormal phenomenon may suggest a mechanism that plasmas are only ignited in different spots other than the orifice region, and drift along the race track to the orifice. This is somewhat similar to findings by Anders [67,68]. His group has found that during HiPIMS discharge, localized hot ionization zones are actually formed and drifting along the race track (Figure 4.32). The formation of these hot ionization zones was observed with the current density higher than 5.2 A/cm\(^2\). This is estimated based on the conditions used his paper. Lower than this current density, the localization is less prominent and only longer strips of slightly hotter plasma or continuous plasma are seen. They also estimated the drifting speed of the hot ionization zones using Equ. 4.5, which is about \( 10^4 \) m/s, lower than the typical \( \text{ExB} \) drifting speed for electrons of about \( 10^5 \) m/s. In the equation, \( B \) is the magnetic flux density, \( Q \) is the charge of ions, \( V_{\text{sheath}} \) is sheath potential drop, \( E_{\text{mps}} \) is the estimated electric field in the magnetic presheath, \( d \) is the distance of ionization location from the target surface, \( m_e \) and \( m_i \) are the electron and ion mass respectively.

\[
v_{IZ} = \frac{2}{B} \left( \frac{Q V_{\text{sheath}} E_{\text{mps}}}{d} \right)^{1/2} \frac{m_e}{m_i}
\]

Figure 4.32 Localized ionization zones (spots of intense plasma) drifting during HiPIMS, as reported by Anders, et. al [68].
Nevertheless, there are some obvious differences in our discharge conditions. In our configuration, the B field is not the same along the race track. So it can be imagined that the plasma is always ignited where the B field is strong and drifts toward the weak-B region in which the orifice is located. Our average current density is estimated to be about 2.9 A/cm$^2$, a little lower than the above calculated value for the formation of localized hot plasma zones. But the current density in the strong B field region should be higher and therefore the plasma over there may have developed into localized hot spots or at least a long strip.

A further calculation of the drifting speed of the plasma is done for the tested recipes. The Equ. 4.5 is used but the values of different parameters are replaced with our own when applicable. For example, B is about 500 Gauss, $V_{\text{sheath}}$ is 350, 400, and 450 for the 750, 800, and 900 V recipes based on measurements, and d is 5 mm based on the observation of bright plasma region of about 1 cm (5 mm is then the distance to the center of the ionization region). Then the drifting speed for the 750, 800, and 900 V recipes are estimated to be $8.6 \times 10^{-3}$, $9.2 \times 10^{-3}$ and $9.7 \times 10^{-3}$ m/s. With the observed delay time, the distance that the plasma has drifted before reaching the orifice can be calculated, which is 26.6, 19.3 and 13.6 cm. Based on these values, the starting positions of the drifting are found, as shown in Figure 4.33. Clearly, the 900 V recipe can ignite the plasma in a larger region that extends deeper into the weak-B region. Based on the almost symmetric magnetic field profile, it is believed that plasma can ignite at the opposite point on the right hand side and as well as in the long strip between the two points. This is supported by the deeper erosion grove in this region.
It is interesting to find out how the plasma strip evolves. If its shape and size stay the same, then it is expected to see a drop in the ion current measured on the grid when this strip passes the orifice. The ion currents are measured in three recipes with different pulse durations, 50, 100, and 150 µs, as shown in Figure 4.34.

Figure 4.33 Estimated plasma ignition zones for HiPIMS discharges using different recipes ($V_{ch}$ between 750 and 900 V). Each white arrow marks the edge of plasma stripe after ignition.

Figure 4.34 Ion current measured on the grid as compared with the discharge current in (a) 50, (c) 100, and (c) 150 µs pulses. $V_{ch}$ 750 V, 5 mTorr.
In the 50 µs pulses, the grid current keeps increasing. In the 100 and 150 µs pulses, the ion current also increases but not as fast as the discharge current does. The deviation happens at about 60 µs. Based on the drifting speed, this point corresponds to the ignition edge at the right hand side. Based on this, it can be concluded that the drifting plasma flow still reflects the shape of initial ignition strip, although the plasma has grown more intense over the entire race track.

### 4.4.2 Differentiation of Various Species

The ion flux measured above contains both Ar⁺ and Cu⁺ ions. It is desirable to distinguish these two species. The first plan is to use the biased QCM to determine the Cu⁺ flux. By applying different biases, the ions are either collected or repelled by the QCM. The difference in the deposition rate will be used to calculate the Cu⁺ flux. Figure 4.35 shows such a set of data with different biases.
applied to the QCM. However, the deposition rates at -500 V and at +100 V are about the same, at much lower values than expected. It is suspected that the pressure in the test chamber may be too high that the gas scattering becomes a serious problem.

![Graph showing deposition rates vs QCM bias](image)

Figure 4.35 Deposition rates measured on the QCM at different bias. Recipe used is 900 V, 50µs, 70 Hz, 5 mTorr.

To test this suspicion, the current collecting plate is placed at different distance from the first grid, as 3 and 10 mm. The current is also measured on the QCM which is 25 mm away. A fixed bias of -300 V is applied to the first grid. The second plate or the QCM is also biased at -300V. As one can see in Figure 4.36 (a), the current dramatically decreases as the distance between the grid and the plate/QCM increases. The ion currents at t = 50µs are extracted and subjected to an exponential decay fit (Figure 4.36 b). Such decay is likely due to a higher pressure than the 5 mTorr in the main chamber due to the slow pumping speed through the very small hole.

![Graph showing ion current vs distance](image)

Figure 4.36 (a) Ion current collected at difference distances from the first grid. (b) An exponential fit of the ion currents collected at the three locations at t = 50µs.

To find an alternative, witness Si wafers are used to measure the metal deposition. The Si
wafers are mounted parallel to the side wall of the isolating ceramic tube. They cover the entire distance between the grid and the QCM, but are kept insulated from them. The recipe of 900 V, 50µs, 100 Hz, 5 mTorr is run for a total time of 70 minutes to deposit thick enough film on the wafer. For the first sample, the electron repeller grid is biased at -500 V, and for the second sample, it is at -50 V to repel the ions. The QCM has been kept at -500 V. Cross-section SEMs are used to measure the deposition thicknesses at different points on the wafer. Figure 4.37 shows the film thicknesses as a function of the distance from the grid. The last point at 25 mm away is from the QCM measurement. It clearly shows the decrease of the deposition thickness, supporting our previous assumption of scattering. Higher deposition thicknesses on the first witness wafer are from the Cu ions. The difference between the two thicknesses is calculated as shown in the same figure. These ions are scattered toward the chamber wall and don’t reach the QCM region. It is seen that the neutral flux seems to saturate at about 20 nm without any further decrease after 11 mm while calculated ion flux reduces to zero in these locations. (The ion current measurements in Figure 4.36 actually show there is still ion flux after 11 mm all the way to the QCM location. Also the equivalent deposition thickness on the QCM can be determined to be 10.7 nm instead of being saturated at 20 nm as shown as the point at about 23 mm.) Two factors should be considered here. First of all, the measurements from the SEM images become less accurate as the thicknesses become lower than 20 nm. Second, there may be still high energy ions that are not repelled by the -50 V on the first grid.
Figure 4.37 Deposition thicknesses on the Si witness plate as a function of the distance to the electron repeller grid. Two different biases -500 V and -50 V are used on the grid. The subtraction of the two is also included.

To further understand such deposition profile observed inside the test chamber, an advanced and extremely useful model developed by John Sporre [87] at CPMI is used. This is a Monte-Carlo simulation used to follow the path traversed by an initialized test atom or ion as it is introduced into a gaseous chamber environment with a preset energy, direction, and location. For the ions, each is created with energy of 400 eV for high energy group or 200 eV for the lower energy group and sent along the Z-direction (from orifice toward the QCM). The initial location of these species is modeled as uniformly distributed on the surface of the orifice opening. The neutrals are modeled using the same distribution but with an initial energy of 3 eV as commonly used for the sputtered atoms. The geometries can be seen in Figure 4.38.

As each species travels through the test chamber, the model tests whether a gas collision or wall collision has occurred. The gas collision cross section is derived using interatomic potentials calculated by an Abrahamson potential coupled with an attractive well. If a collision is deemed to have occurred in a given step length, the collision is then carried out using classical scattering theory. An impact parameter is chosen as $b=b_{\text{max}} a^{0.5}$ where $b_{\text{max}}$ is the value at which scattering is less than 1°, and $a$ is a random number between 0 and 1. Wall collisions are based on the theoretical measurements.
observed using the SRIM scattering analysis code. Ultimately, the SRIM analysis allows for the determination of scattering, deposition, and sputtering probabilities, as well as changes in energy pre and post wall interaction, as a function of incident angle and energy. Once the test atom has deposited, or reached threshold energy of 0.001 eV, the code is stopped and repeated for the next test atom. In total, 1000 test atoms were used for each measurement.

The simulation results of the Cu atom deposition and the fast ion deposition at 100 mTorr are shown in Figure 4.38. The top graph shows the Cu deposition on the side wall at different distances from the orifice opening (the top surface of the test chamber). The grid is located at about 8 mm away as marked in the graph. It can be seen that the neutral deposition on the sidewall quickly decreases from the grid to the QCM (the end of the axis). No obvious deposition occurs for neutral atoms based on simulation. For fast ions, the simulated deposition profile is different. It reaches a maximum at about 10 mm away and remains high for a large range. It also leads to some deposition on the end wall of QCM. Based on our assumption, when the mesh is biased at -50 V, there are still some high energy electrons penetrating through. The measurement on the witness plate is thus added in Figure 4.38. The comparison between the measured thickness and the simulated results show that the above assumption helps explain the saturation after 12 mm. If given the right ratio between the atoms and fast ions, the deposition can definitely be fit using the simulation results.

![Figure 4.38 Neutral atoms deposition and fast ions deposition on the side wall and bottom wall based on the simulation at 100 mTorr. Also shown is the measured deposition thickness on the Si witness wafer along the sidewall. The dashed line shows the equivalent thickness measured on the QCM.](image)

The simulation results of ions with lower energy (200 eV) at 100 mTorr are given in Figure 4.39. It can be seen that the deposition on the sidewall decreases from the grid to the QCM. This
agrees with the ions measured from the subtraction. The lower energy than the target potential is possible. First, ions may be generated at different distance from the target in the plasma. The plasma potential varies with this distance, which leaves ions with different energy after being accelerated and introduced into the hole. Second, ions very likely will have sputtering inside the hole to further lose energy.

![Figure 4.39 Distribution of 200 eV ions deposition based on the simulation at 100 mTorr.](image)

Going back to Figure 4.37, the deposition flux of Cu atoms and Cu ions are calculated by integrating the deposition thickness over the entire witness plate. It can be determined that there are about 40% of Cu ions in total Cu flux. Comparing the total Cu⁺ ions flux and the average ion current collected on grid, it can be determined the ratio of Cu⁺ to Ar⁺ is about 55%. So the through-target flux measurement confirms that self-sputtering becomes important in HiPIMS of Cu. More discussion of the Cu⁺ to Ar⁺ ratio will be done combined with a plasma model in the next Chapter.

### 4.5 Conclusions

One of the motivations of this work is to evaluate HiPIMS as a potential technique to further extend iPVD for barrier/seed layer deposition. Different from the existing HiPIMS applications, the barrier/seed deposition processes have very stringent requirements on the plasma environments, such as plasma uniformity, metal ion fraction, etc. Detailed characterizations of the HiPIMS plasma are thus performed in the beginning to find out its performance. Effects of different pulsing and discharge parameters are studied.

Very high peak current up to 750 A can be achieved. High electron densities ($n_e$) during the
pulse to about \(5 \times 10^{17} \text{ m}^{-3}\) are generated on the substrate and reach \(3 \times 10^{18} \text{ m}^{-3}\) later after the pulse ends. Cu ionization fractions (IF) are measured on the substrate level. Up to 60% has been achieved using a 200 Gauss magnetic field configuration, much higher than the DC magnetron sputtering. It basically increases with higher charging voltage and longer pulse length due to higher plasma densities. A stronger B field, however, produces lower Cu ionization in spite of its higher plasma density. This is attributed to a lower ion extraction efficiency due to the plasma potential distribution.

During the study, some distinctive features have been observed. Their possible effects on the application, as well as the underlying physics are further investigated. Plasma expansion is observed with a high plasma density peak moving from the target and arriving at the substrate about 150 µs later. Faster expansion is observed with a higher charging voltage or a longer pulse duration. It is believed that a high enough \(n_e\) in the confined plasma leads to an enhanced plasma diffusion across the magnetic field. The higher is the \(n_e\), the earlier the diffusion starts, and the faster the expansion. The magnetic field also strongly affects the plasma transport. With an increased magnetic field strength from 200 Gauss to 800 Gauss, the plasma expansion becomes faster and more directional towards the substrate as oppose to nearly isotropic in 200 Gauss. A large electric potential drop is observed in the presheath and extends into the bulk plasma region. This potential drop remains large for a long time in the 800 Gauss configuration. The corresponding electric field promotes the electron drifting. At the same time, the higher density in the 800 Gauss configuration promotes the diffusion process across the B field. Both lead to a faster plasma expansion.

Plasma fluxes to the cathode are for the first time directly measured through an orifice in the race track region. The ion fluxes are measured on a negatively biased grid. It is seen that the ion current increases abruptly after a certain time delay instead of increasing smoothly from the pulse beginning. This time delay depends on recipes, for example, being longer for lower charging voltages. This suggests a mechanism that plasma is ignited only in a long strip where the B field is strong and drifts toward the weak-B region. A higher charging voltage allows plasma to ignite in a larger region and drift faster. Fluxes of Cu atoms and Cu\(^+\) ions are measured using Si witness plates and then compared with the total ion flux. A large ratio of Cu\(^+\) to Ar\(^+\) is determined as a direct evidence for the enhanced self-sputtering during HiPIMS.

In all, the plasma characterizations show overall advantages of HiPIMS over normal DC
sputtering. However, the HiPIMS discharge is found to be much more complex than the steady-state DC discharge. For example, the HiPIMS plasma expansion after the pulse may open up for new possibilities of an independent control and thus more flexibility for the HiPIMS application. It should be paid attention to though, because the 3D plasma transport study reveals a change of expansion orientation from isotropic to directional, which will dramatically affect the deposition uniformity on the substrate. A careful design of the magnetic field may be necessary. Other than this, another HiPIMS discharge mechanism being studied in this chapter is the plasma potential distribution and its effect. It obviously affect the plasma expansion speed and orientation, and also the ion extraction efficiency, which will be critical for the interconnect metallization application. One special contribution here is the through-target flux diagnostics. It is the first time that direct evidences are presented for the self-sputtering effect. The diagnostics can certainly be applied to various conditions to reveal more physics involving the HiPIMS plasma generation.
Chapter 5  HiPIMS Ionization Region Model

HiPIMS has been shown to have very complicated and unique mechanisms, many of which are still unclear. A lot of efforts have been devoted to HiPIMS plasma modeling by different researchers. A time-dependent model is usually necessary to study the HiPIMS discharge mechanisms. In this chapter, a simple time-dependent model is built to describe the ionization reactions inside the strong magnetic-field-confined plasma. Using some experimental data as the input, the model is able to predict the temporal densities of different species including electrons, metal atoms, and ions of both metal and argon. The model also depicts some essential processes during the HiPIMS discharge.

5.1 Model Description

HiPIMS plasma modeling has been developed for many years, and has been proved helpful in understanding the complex HiPIMS plasma discharge [70,88,89]. Their work has been learned and used as good references for the our own model development. The model is built to describe the ionization zone in vicinity of the target. It is known that the magnetic field defines a region with an effective plasma confinement. Ionizations are greatly enhanced, leading to the erosion race track formation in front of this strong plasma region. The geometry of the ionization zone is described in a schematic diagram in Figure 5.1. For the modeling simplicity, it can be assumed to have a rectangular cross section. The lateral size is taken as same as the race track width w. In axial direction, it is between $z_1$ and $z_2$. Here $z_1$ is the sheath width, while $z_2$ is determined by the magnetic field topology. In this model, although there is no direct input of B field, it does affect the results through the choice of dimensions for the ionization region. Typically $z_1$ value is quite small (e.g., 1 or 2 mm), while $z_2$ is between 1 and 3 cm from literature [88]. In our experiments, by observing the brightness of the plasma, $z_2$ is about 1.5 cm.

The species to be considered in the model include the electrons, metal atoms $M^0$ and ions $M^+$, Ar atoms $Ar^0$ and $Ar^+$ ions. Electrons are emitted from the target and gain energy from the sheath. It loses the energy through excitations and ionizations. The reaction rates depend on the electron energies in the plasma. In a Maxwellian-distributed plasma, it is common to use electron temperature $T_e$ instead. The created ions are then accelerated toward the target and induce sputtering in the race
track. The ejected metal atoms will traverse through the plasma and get partially ionized. The degree of ionization again depends on the instantaneous plasma properties.

![Figure 5.1 A schematic diagram showing the ionization region.](image)

Figure 5.2 shows a block diagram illustrating the structure of the model. Before getting to the detailed processes and corresponding equations, the input parameters are briefly described. The discharge voltage $U_d(t)$, current $I_d(t)$ and pressure $P$ can be easily obtained from experimental measurements. These voltage and current waveforms are not independent. Technically, by just using $U_d(t)$ as the input, the entire HiPIMS discharge can be predicted including the discharge current. However, the model will be much more complicated. $U_d$ affect the important processes on the target such as sputtering via the dependence of sputtering yield on the incident ion energies. A large fraction of $U_d$ is dropped in the sheath region to accelerate the ions. Therefore in this model, the sheath voltage and thus the ion energies are taken to be equal to $U_d$. The sputtering yield of Cu by both $Ar^+$ and $Cu^+$ at a perpendicular incident angle at different energies are calculated and fitted, as shown in Figure 5.3. The discharge voltage also affects the secondary electron emission and their energies after sheath acceleration. This process, however, is not necessary to consider in the current model since the discharge current $I_d(t)$ is also used as the input. $I_d(t)$ include both the $Ar^+$ and $Cu^+$ components, whose relative ratio varies over time and is calculated in the model.

The geometry of the ionization region is also used as the model input. As briefly mentioned above, the race track width $w$ is measured and used as the width of the ionization zone. 1 mm is used for the sheath width $z_1$. The height $z_2$ is chosen to be 1.5 cm for initial tests based on plasma
observations. Uniform $n_e$ and $T_e$ are used in this region. The Cu atom density varies along the axial direction considering several microseconds are needed for Cu atom flux to pass the ionization zone. The front and the end surfaces thus see the Cu atom fluxes generated on the target by several microseconds apart. An average Cu atom density in the region is then used.

$T_e$ is another model input which can be predicted based on the experimental measurements. This eliminates the need for including all the loss mechanisms in the power balance equation.

![Block Diagram of the Model](image)

**Figure 5.2** A block diagram showing the structure of the model.

![Sputtering Yield vs Ion Energy](image)

**Figure 5.3** The Cu sputtering yield $Y$ as a function of ion incidence energy for both (left) Ar$^+$ and (right) Cu$^+$. 

In the model, densities of different species including $n_e$, $n_{Cu}^0$, $n_{Ar}^0$, $n_{Cu}^+$, $n_{Ar}^+$ are calculated at the beginning of each time step. With increased $n_e$ (from increased $I_d$) and $n_{Cu}^0$ (from more intense...
sputtering), new $n_{\text{Cu}}^+$ and $n_{\text{Ar}}^+$ are achieved at the end of each time step. A new ratio of $n_{\text{Cu}}^+$ to $n_{\text{Ar}}^+$ is then used to determine the $n_{\text{Cu}}^+$ and $n_{\text{Ar}}^+$ for the next step.

The details of the calculations are given below.

1) Prediction of $n_{\text{Cu}}^+$, $n_{\text{Ar}}^+$ and $n_e$. The discharge current $I_d$ is composed of $I_{\text{Ar}}^+$ and $I_{\text{Cu}}^+$ (Equ. 5.1). At the initial time, there is only Ar$^0$ in the chamber, so the plasma contains only electrons and Ar$^+$. The density of Ar$^+$ $n_{\text{Ar}}^+$ can then be determined using (Equ. 5.2). This is based on the mechanism that ions are accelerated to Bohm velocity at the sheath-presheath edge and the flux remains the same in the sheath. Similar calculations can be done for Cu$^+$ using Equ. 5.3. The Bohm velocity depends on the electron temperature $T_e$ and the mass of either Cu$^+$ or Ar$^+$ ions (Equ. 5.4). Other parameters in these equations include $e$ as the elementary charge, $A$ as the race track area and $M$ as the ions. Electron density is just calculated as the sum of $n_{\text{Cu}}^+$ and $n_{\text{Ar}}^+$, based on the charge neutrality in the plasma.

\[
I_d = I_{\text{Ar}}^+ + I_{\text{Cu}}^+ \\
I_{\text{Ar}}^+ = n_{\text{Ar}}^+ e u_B (\text{Ar}^+) A \\
I_{\text{Cu}}^+ = n_{\text{Cu}}^+ e u_B (\text{Cu}^+) A \\
u_B = \left(\frac{e T_e}{M}\right)^{\frac{1}{2}} \\
n_e = n_{\text{Ar}}^+ + n_{\text{Cu}}^+ \\
5.1 \\
5.2 \\
5.3 \\
5.4 \\
5.5
\]

2) Prediction of $n_{\text{Cu}(0)}$ from sputtering. The Cu target is sputtered by both Ar and Cu ions, with their own sputtering yields, as shown in Equ. 5.6. These Cu atoms have an average energy of between 2 and 3 eV leaving the target, based on which an average speed can be calculated as $v_0$. At 2 eV, the Cu atoms have a speed of about $1.7 \times 10^3$ m/s. The density of Cu atoms can then be determined using Equ. 5.6 It should be noted that with this speed, Cu atoms need about 8.6 $\mu$s to penetrate the ionization zone of 1.5 cm. As a result, the Cu atom density in the region is not uniform even when no scattering collisions take place. An average Cu atom density in the region is then calculated for any short time steps.

\[
\Gamma_{\text{Cu}(0)} = \Gamma_{\text{Ar}}^+ Y_{\text{Ar}}^+ + \Gamma_{\text{Cu}}^+ Y_{\text{Cu}}^+ \\
\Gamma_{\text{Cu}(0)} = n_0 v_0 \\
5.6 \\
5.7
\]
3) Rarefaction effect. During HiPIMS, the fast sputtered metal atoms will have collisions with the background Ar atoms. This “sputtering wind” thus reduces the argon density. An accurate prediction of the argon density loss requires the knowledge of the densities of different species including Cu⁰ and Cu⁺ as well as their velocity distributions, based on which a detailed calculation of momentum transfer can be made. In the model, we use a simplified way to deal with this effect. The argon density is reduced to keep the pressure (or the total density) in the region constant.

4) Ionization processes. The ionization rate is described by Eqn. 5.8. It is proportional to both the neutral and electron densities. Here, $k_{iz}$ is the ionization rate coefficient based on the ionization cross sections and the electron energy distributions. Both the electron impact ionization and Penning ionization are considered with the cross-sections taken from literatures [88]. The energy distribution is assumed to be Maxwellian for the calculation of $k_{iz}$. Other than the ion generation in the ionization region, the ion loss is also included by subtracting the target-directed ion flux, as shown in Eqn. 5.10. A new $n_{Cu}^+$ is calculated for each short time step. Similar calculation is done for Ar ions at the same time.

\[
\frac{d n_{Cu}^+}{dt} = n_0 n_e k_{iz} \tag{5.8}
\]

\[
k_{iz} = \langle \sigma_{iz} v_e \rangle \tag{5.9}
\]

\[
V \frac{dn_{Cu}^+}{dt} = V n_0 n_e k_{iz} - n_{Cu}^+ e u_B (Cu^+) A \tag{5.10}
\]

5) Time development. With a newly calculated $n_{Cu}^+$ and $n_{Ar}^+$, technically the model will continue to run without the need for further input. But in fact, we do have the $I_d(t)$ data. To avoid inconsistency, only the ratio of the $n_{Cu}^+$ and $n_{Ar}^+$ is kept for the next time step. New $I_{Ar}^+$ and $I_{Cu}^+$ are determined to continue the calculation.

5.2 Results and Discussion

The model is first tested using the data taken from a typical HiPIMS discharge using the 500G magnetron configuration with Cu target. A recipe of 800 V, 50 µs, 100 Hz, 5 mTorr in Ar has been extensively characterized and is used here. It has a ring-shaped race track of 11.7 cm in radius and 2.8
cm in width. The electron temperature is measured to be about 1.5 eV during the pulse at \( z = 1 \) cm, and will be used for the model. Again \( z_1 \) and \( z_2 \) are chosen to be 1 mm and 1.5 cm respectively. The input voltage and current waveforms are provided in Figure 5.4 for reference.

![Figure 5.4 Model input of \( U_d(t) \) and \( I_d(t) \) from the experimental measurement. Recipe is 800 V, 50 \( \mu \)s, 100 Hz, 5 mTorr in Ar.](image)

The model results without including the rarefaction effect are shown in Figure 5.5. The densities of electrons, \( \text{Ar}^+ \) and \( \text{Cu}^+ \) ions are shown in Figure 5.5 (a). The input discharge current is shown for time reference. Significant increase of \( n_e \) up to \( 6 \times 10^{19} \) m\(^{-3} \) is shown. Such a high density has been measured using triple Langmuir probe previously. The \( \text{Ar}^+ \) density \( n_{\text{Ar}^+} \) gradually increases during the pulse, as a result of the intensifying plasma. The \( \text{Cu}^+ \) density \( n_{\text{Cu}^+} \) which is zero at the beginning rises more quickly and surpasses \( n_{\text{Ar}^+} \) close to the end of the pulse. This is due to an

![Figure 5.5 Model results for 800 V, 50 \( \mu \)s, 100 Hz, 5 mTorr without the consideration of rarefaction. (a) The electron density and the densities of \( \text{Ar}^+ \) and \( \text{Cu}^+ \). (b) The ionization fraction of \( \text{Cu} \), and of argon, and the fraction of \( \text{Cu}^+ \) ions in total ions. In both figures, the input discharge current is shown for time reference.](image)
increasing accumulated Cu neutral density. Figure 5.5 (b) further shows the ionization fractions of Cu and of argon, and the fraction of Cu\(^+\) ions in total ions. The Cu IF increases to 27\% and the Ar IF increases to 17\%. The fraction of Cu\(^+\) ions in total ions reaches 54\% before the pulse ends.

The model is then run taking account of Ar rarefaction effect. The results are shown in Figure 5.6. One change is the noticeable decrease of Ar\(^+\) density. This is obviously originated from a continuously decreasing Ar neutral density (not shown). For the same reason, Cu\(^+\) ions become more dominant in the total ions, reaching 67\% as compared with 54\% without including rarefaction. The Cu degrees of ionization for Cu and Ar are about 32\% and 26\%. Since the rarefaction effect considerably changes the model results, it is included in all the subsequent model runs.

![Figure 5.6 Model results for 800 V, 50 \(\mu\)s, 100 Hz, 5 mTorr with rarefaction effect. (a) The electron density and the densities of Ar\(^+\) and Cu\(^+\). (b) The ionization fraction of Cu, and of argon, and the fraction of Cu\(^+\) ions in total ions.](image)

The height of the ionization region \(z_2\) as the input parameter is changed from 1.5 to 2 cm to test the sensitivity of model on this parameter is tested. The results are shown in Figure 5.6. It can be seen that the change of \(z_2\) does not significantly alter the model results. This is expected since the model is essentially a zero-dimensional that the plasma inside the ionization region is uniform.
Figure 5.7 Model results using different $z_2$ input for 800 V, 50 µs, 100 Hz, 5 mTorr with rarefaction effect. (a) The electron density and the densities of Ar$^+$ and Cu$^+$. (b) The ionization fraction of Cu, and of argon, and the fraction of Cu$^+$ ions in total ions.

Another input parameter $T_e$ is varied to see its effect on the model. 2 eV is used as opposed to 1.5 eV used above. It should be noted in advance that for a specific discharge, $T_e$ should be fixed. Using a different value will cause some inconsistency. The results are shown in Figure 5.8. Greatly increased $n_{Cu^+}$ and ratio of $n_{Cu^+}/n_e$ are seen. This is because the Cu ionization rate has increased more rapidly than the Ar ionization. One obvious discrepancy is between the lower $n_{Ar^+}$ observed and increased Ar ionization rate at higher $T_e$. By choosing a higher $T_e$, both $n_{Ar^+}$ and $n_{Cu^+}$ increase, but $n_e$ is actually constrained by the same $I_d$ input. In a word, $T_e$ can considerately affect the model results, especially the ratio of Cu$^+$ to Ar$^+$. The choice of $T_e$ has to be careful, like in our case with the assistance of experimental measurements. Otherwise, a global model including the power balance is needed to predict the $T_e$ during discharge.

Figure 5.8 Model results using different $T_e$ input for 800 V, 50 µs, 100 Hz, 5 mTorr with rarefaction effect. (a) The electron density and the densities of Ar$^+$ and Cu$^+$. (b) The ionization fraction of Cu, and of argon, and the fraction of Cu$^+$ ions in total ions.
The different discharges can then be modeled. The effects of different parameters such as charging voltage, pulse duration, B field can be studied. An example is given using a recipe with higher $V_{ch}$ (900 V, 50 µs, 100 Hz, 5 mTorr). The results are shown in Figure 5.9. If compared with the 800V recipe (Figure 5.6), it can be seen that both Cu$^+$ density and Ar$^+$ density increase. More interestingly, both Cu ionization fraction and Ar ionization fraction increase to about 40%. The model thus can be used as a method to provide basic ideas of a recipe’s performance even before plasma diagnostics.

![Figure 5.9 Model results for 900 V, 50 µs, 100 Hz, 5 mTorr with rarefaction effect. (a) The electron density and the densities of Ar$^+$ and Cu$^+$. (b) The ionization fraction of Cu, and of argon, and the fraction of Cu$^+$ ions in total ions.](image)

Finally, the discharge used in the through-target flux measurement (Section 4.4) is modeled. The old horseshoe-shaped magnetic field configuration and 900 V, 50 µs recipes were used in 5 mTorr Ar. The discharge voltage and current waveforms are given in Figure 5.10. Other input parameters are $z_1 = 1$ mm, $z_2 = 1.5$ cm, and $T_e = 1.5$ eV. Racetrack area is measured to be 422.8 cm$^2$.

![Figure 5.10 $U_d(t)$ and $I_d(t)$ from the experimental measurement. Recipe is 900 V, 50 µs, 100 Hz, 5 mTorr in Ar.](image)
The model results are shown in Figure 5.11. Electron density is seen to increase up to 6.6×10^{19} \text{ m}^{-3}. Since the magnetic field is not symmetric in this configuration, so that \( n_e \) varies at different points in the race track. The \( n_{\text{Ar}^+} \) increases at the beginning but decreases later due to the gas rarefaction. The \( \text{Cu}^+ \) density \( n_{\text{Cu}^+} \) increases more rapidly and becomes dominant after 30 \( \mu \text{s} \). This is due to an increasing Cu neutral density. The Cu IF increases to 33\% and the Ar IF increases to 21\%. Such values are lower than what to be expected from the HiPIMS discharge. On one hand, this may be because the \( T_e \) we choose is lower than the real values. As a consequence, the ionization cross-sections are underestimated. On the other hand, this is more likely caused by the not-high-enough current density. Due to the large size of our target, it is relatively difficult to apply a current of 1000 V or higher while keeping the discharge stable. This leads to a relatively low \( n_e \) for further enhancement of the ionization.

![Figure 5.11](image)

Figure 5.11 Model results. (a) The electron density and the densities of Ar\(^+\) and Cu\(^+\). (b) The ionization fraction of Cu, and of argon, and the fraction of Cu\(^+\) ions in total ions

The fraction of Cu\(^+\) ions in total ions reaches 82\% before the pulse ends. So it is almost completely self-sputtering at this point. In the through-target diagnosis, we have measured the ratio of Cu\(^+\) to Ar\(^+\) as 0.55. This is a result derived from the witness plate deposition tests, so it is an average value over time. For comparison, the time-averaged value of \( n_{\text{Cu}^+}/n_{\text{Ar}^+} \) ratio from \( t = 0 \) to \( t = 50 \mu \text{s} \) from the model is 1.58. This is likely because the orifice is located in the weak B field region. The plasma density above it is substantially lower than the strong B field region. The model based on a symmetric B field thus overestimates the plasma density (thus the ionization capability), the flux of sputtered Cu atoms, and the gas rarefaction. Nevertheless, both the experiments and the model show the self-sputtering plays an important role in the HiPIMS discharge.
The model is shown to be useful to quickly predict some key parameters in HiPIMS and to help understand the important processes such as the self-sputtering. The time-dependent electron, ion and neutral density are determined. Based on these values, the degrees of gas rarefaction and self-sputtering can be estimated. This model can be especially useful for practical use when diagnostics are costly and time-consuming to implement, with the discharge voltage and current usually easy to obtain. The model can be further expanded to include the power balance and the particle loss, so that electron temperature can be calculated to provide more accurate results. If the potential distribution is included, the ion extraction efficiency can be estimated.

This work on model also proves the overall importance of plasma modeling in understanding the plasma discharge, especially in a complex discharge as in HiPIMS. Appropriate assumptions are important to simplify the processes while highlighting the key mechanisms.
Chapter 6  Optimization of Magnetic Field Configuration

TLP Langmuir probe measurements have shown that the magnetic field strength has significant influences on the pulsed plasma generation and transport. The extremely dense plasma may lead to a preferred orientation for the expansion and consequently affect the uniformity of the downstream plasma. It is also desirable to have a full face target erosion to control the re-deposition. It would be advantageous to have new designs of magnetron configurations with the more uniform plasma distribution even near the target while maintaining the high pulse current. Different designs have been created and further tested for both IV characteristics and the plasma distributions.

6.1 Interconnected Racetracks

For better target utilization and better deposition uniformity, more evenly distributed race tracks on the target are desired. First group of designs use many groups of magnets across the whole surface with the polarities opposite to the adjacent groups. Figure 6.1 shows two designs (Interconnected No. 1 and Interconnected No. 2) and their corresponding $B_{//}$ at the target surface. The $B_{//}$ forms a number of circles to have more uniform coverage on the target, instead of a single ring as seen in the previous chapter. However, this is a non-closed configuration. The behaviors of electrons at the connection point of two neighboring circles need to be studied, i.e. whether they stay drifting in the same circle or hop from one circle to the next or escape from confinement.

However, the discharge tests revealed a problem. Plasma leaks out from certain points and the discharge current is quite low. Figure 6.2 shows some of the leaking point as pointed by the arrows. At these three points, the $E \times B$ drift of electrons has an outgoing direction. While at the other three locations where $E \times B$ drift points inward, there is no sign of plasma leak. It is also believed that plasma can leak from the connection point of two neighboring circles where magnetic cusp is formed.
Figure 6.1 Interconnected Design No. 1 (left) and No.2 (right) to have interconnected race tracks. The bottom two figures show the corresponding $B_x$ profiles on the target surface.

Figure 6.2 Race tracks showing the leaking points due to non-closed magnetic field. No. 1 (left) and No.2 (right).
6.2 Circular Designs

Circular-shaped designs were made, while trying to include as long race tracks as possible. The first design in this group has three loops of magnets but each of them is divided into halves with opposite polarities, as shown in Figure 6.3 (a). One of the obvious benefits of this design is the capability of sputtering the center of the target. The concern is the magnet polarities are switched at multiple points, forming magnetic cusps and likely non-closed race tracks. The configuration is named “Circular design No. 1”.

Figure 6.3 Circular design No. 1. (a) The magnet arrangements. (b) The $B_{//}$ profile on the target surface. (c) The typical discharge current waveforms. (d) Leak point of the plasma.

The discharge tests were then performed, which showed very low pulse currents being
generated (Figure 6.3 c). Only up to 15 A was achieved as compared to several hundred A from the previous configurations. And obvious leak of the plasma was spotted as shown in Figure 6.3 (d). Analyzing the E×B drift path of electrons (as marked in Figure 6.3 b), it can be seen electrons drift along the path on the entire surface but eventually leak out from the point on the right. This greatly reduced the discharge current.

![Diagram](image1.png)  
**Figure 6.4 Circular design No. 2.** (a) The magnet arrangements. (b) The B_{||} profile on the target surface. (c) The typical discharge current waveforms. (d) Leak point of the plasma.

A modification was then made based on Circular design No. 1. Near the leak point, a row of magnets of north polarity was added to extend into the south polarity magnets, with the hope to block the E×B drift path going out. The configuration is named as Circular design No. 2, shown in Figure 6.4 (a).

The discharge currents were seen to increase as compared with No. 1, but they were still less than 100 A. The sputtering race track revealed the problem that even though the previous leaking path was blocked and the plasma generated in the top half the race tracks can be well confined, a new
leaking path was formed at the bottom part of the design. This is illustrated in Figure 6.4 (b).

### 6.3 Closed Race Track Designs

All the above attempts have one common problem that the drift path of the electrons is not closed. As results, the plasma can leak out from one or more points. So from now on, designs are made to have closed drifting paths.

![Closed Race Track Designs](image)

**Figure 6.5 Closed No. 1.** (a) The magnet arrangements. (b) The B\_ profile on the target surface. (c) The typical discharge current waveforms. (d) Discharge current with and without magnet rotation.

Figure 6.5 shows a new design, “Closed No.1”. It has a complete outer circle to avoid any open loop. The inner arrangement of magnets is similar to the above so that the center of the target can also be sputtered. The B field distribution is shown in Figure 6.5 (b). A possible leaking point due
to magnetic cusp is seen at the lower right part. The discharge tests were then performed. Higher current were achieved but were still lower than those in 500G which has a simple ring-shaped race track and similar B field strength. One interesting observation is that when the magnet rotated, the discharge current greatly increased, as shown in Figure 6.5 (d). Obviously, at the weak point the electrons trying to escape the confinement by diffusing perpendicular to the target will be re-captured by the spinning stronger magnetic field.

![Magnet Arrangements](image1.png)

**Figure 6.6 Closed No. 2.** (a) The magnet arrangements. (b) The B∥ profile on the target surface. (c) The typical discharge current waveforms. (d) Race track marks.

The lesson learned from the above design is no magnetic cusp should be allowed. Based on this, Closed No. 2 design was then made as shown in Figure 6.6. Two separate but closed race tracks are expected to form based on the B field profile. The only possible weak point is marked in figure (b) as the dashed line. The discharge current was shown to be quite high. The plasma was also very bright,
however, plasma was only observed in the outer ring race track. On the target, there is only very weak mark of sputtering at the central part. Very likely the weak $B_\parallel$ point in the design caused the issue.

To order to have both high discharge current and long race tracks, a spiral-shaped design was made. Its design and the corresponding $B_\parallel$ profile are shown in Figure 6.7. It also has more coverage of race tracks on the target as one can see in Figure 6.7 (c). Its discharge current was measured to be comparable with 500G configuration, as shown in Figure 6.9.

![Figure 6.7 Spiral-shaped design (a) The magnet arrangements. (b) The $B_\parallel$ profile on the target surface. (c) The typical plasma during discharge. The arrow in (b) shows the radial orientation of 3-D triple probe measurement as described in the following section.](image)

Designs were then made to change the race track width while keeping a similar maximum $B_\parallel$. 500G configuration was used as a baseline. By moving the inner circle of magnets further inward and adjusting the number density of the magnets, the maximum $B_\parallel$ was maintained at about 500 Gauss. Meanwhile, the strong field region (e.g. of $B_\parallel$ greater than 200 Gauss) spanned about twice wider than that in the 500G baseline configuration, as shown in Figure 6.8 (b). A wider race track was indeed created as confirmed from the subsequent observance of the discharge plasma. This field configuration is later referred as “500G_wide”. Its discharge current was found to be about twice higher than that in the 500G configuration, as shown in Figure 6.9. This is believed to be originated from its almost twice wider race track.
Figure 6.8 The magnet arrangements (top row) and the corresponding $B_y$ on the target surface (bottom row) in different configurations, (a) 500G, (b) 500G-wide.

Figure 6.9 Discharge characteristics for three configurations, baseline 500G, 500G-wide, and Spiral, using 800 V, 50µs, 100 Hz, 5 mTorr.
Both the 500G_wide configuration and spiral configuration have shown certain benefits to the target utilization, either with wider or longer race track. Triple Langmuir probe was then used to map the plasma distribution to determine the uniformity. Figure 6.10 compares the peak \( n_e \) and the peak delay time in the 500G, 500G_wide and spiral configurations. For 500G_wide case, a higher peak \( n_e \) was obtained near the race track, consistent with the higher discharge current measured. A hump of \( n_e \) over an extended region between \( R = 4 \) cm and \( R = 12 \) cm can be observed at different \( Z \) positions. Meanwhile, the peak delay time was shorter in this region. The widened race track indeed spread out the coverage of intense plasma but did not address the low plasma density at the center and the edge of the chamber.

The spiral magnetic field design included multiple turns of race tracks, covering the center as well as the outer edge. Figure 6.10 (c) shows the peak \( n_e \) and the peak delay time using this design. The radial scanning orientation was marked by an arrow in Figure 6.10 (c). Right below the target (\( Z = 13 \) cm), there were three \( n_e \) peaks resulted from the race tracks at \( R = 0, 8, 13 \) cm. The distribution
of the downstream plasma density quickly flattened. The density decrease near the edge was expected since the chamber wall served as a plasma sink. The distribution of the peak delay time was uniform from the center to \( R = 10 \text{ cm} \) on the substrate level. In short, the spiral magnet configuration obtained a plasma with superior uniformity, in addition to its better target utilization. The design can be easily scaled up further for larger-area depositions.

The superiority of the spiral configuration can be more visually seen with the color mapping of the peak \( n_e \) and the peak delay time, as shown in Figure 6.11.

![Figure 6.11 Color mapping of the peak \( n_e \) (top row) and the corresponding peak delay time (bottom row) of the plasma expansion in \( 500G\text{wide} \) and \( \text{Spiral} \) configurations.](image)
6.4 Conclusions

The work in this chapter involves the efforts to optimize the magnetic field configuration specifically for HiPIMS discharge. This has been overlooked for many years of HiPIMS study and applications. The importance of the magnetic field configuration in HiPIMS arises from the non-flat plasma potential distribution associated with the short pulse duration which will affect the plasma expansion orientation. In another word, even with the magnet rotation, the downstream plasma in HiPIMS may reflect more the race track shape than in DC discharge. The uniformity of the process will be affected.

Magnetic field configurations are modified specifically for the high power impulse magnetron sputtering (HIPIMS). The race track pattern is varied to optimize the target utilization and the downstream plasma uniformity. Attempts are made to spread the erosion evenly on the target, but the discharge currents are relatively low due to the non-closed race tracks. A closed path for electrons to drift along is still essential in HIPIMS. The configuration of wider race track generates a higher pulse current, and extends the intense plasma coverage on the substrate. A spiral-shaped magnetic field configuration is able to generate high pulse current, achieve a downstream plasma with superior uniformity, and yield a better target utilization even without the assistance of magnet rotation.
Chapter 7  Study of MPP in Planar Magnetron

Modulated pulsed power (MPP) magnetron sputtering is a new derivative of the HiPIMS that may allow unprecedented user control over the growth process. It has some distinctive features, such as flexible control over the discharge voltage and current waveforms. However, the critical time-dependent plasma properties during the pulse have not been studied. In this chapter, a thorough characterization of the MPP discharge is performed in the Galaxy planar magnetron. The discharge IV characteristics, time resolved plasma parameters ($T_e$, $n_e$, $V_f$), and metal ion fractions are measured under various pulsing and discharge parameters. The effect of the magnetic field on the MPP discharge is also investigated. The MPP can be operated in two different modes, Solo and Cyprium, both of which are studied.

7.1 MPP Solo

MPP has also been studied in Galaxy planar magnetron system using the Al target. The tests being done are listed in Table 7.1. Different from the Huettinger generator, Zpulser MPP plasma generator uses different pulse profiles as described in the previous section. It can also be operated in two different modes in which either repetition frequency or average power is specified. Again, for each test, only results from one or two varied parameters are shown as examples.

| Table 7.1 Tests of MPP sputtering in Galaxy planar magnetron |
|------------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
|                  | Charging voltage | Micropulse on- and off-time | Macropulse length | Repetition frequency/power | Pressure | Various locations |
| I-V characteristics | x                | x                      | x                | x                            | x        | x                |
| Triple probe      | x                | x                      | x                | x                            | x        | x                |
| Ionization fraction (metal) | x          | x                        | x                | x                             | x        |                  |
| Deposition rate   | x                | x                        |                   | x                            | x        | x                |
7.1.1 Discharge I-V Characteristics

The I-V discharge characteristics of the MPP generator are studied prior to the triple probe measurements. The typical I-V curves of the MPP discharge show multiple stages, as seen in Figure 7.1. The macro-pulse is programmed to be 3000 μs long, consisting of micro-pulses of $\tau_{\text{off}}$ 40 μs and $\tau_{\text{on}}$ 6 μs (or shorted as 40-6) in the initial 1500 μs and a session of micro-pulses to gradually ramp up. The weak recipes at the beginning make sure the discharge can be ignited. The main part of the macro-pulse is the last 1000 μs, consisting of the micro-pulses of $\tau_{\text{off}}$ 10 μs and $\tau_{\text{on}}$ 6 to 9 μs. The charging voltage is 450 V and pressure is 5 mTorr. Plasma ignition occurs in the first 100 μs ending with a small surge of current. Then it goes into a low power sputtering mode similar to the conventional DC magnetron sputtering. After the session of higher-duty-ratio micro-pulses starts, the current is dramatically ramped up and may enter the steady state given enough time. In this last stage, the current is very high to effectively ionize the sputtered material.

![Figure 7.1 Typical I-V curves of the MPP discharge, under different $\tau_{\text{on}}$ for the micropulses Discharge parameters are $\tau_{\text{off}} = 10$ μs, $\tau_{\text{on}} = 6-9$ μs, Vch 450 V, and 5 mTorr.](image)

With a constant off-time $\tau_{\text{off}}$ of 10 μs, changing the on-time $\tau_{\text{on}}$ from 6 to 9 μs increases the pulse current from 100 to about 400 A. The voltage, however, doesn’t show obvious difference for the three recipes during the pulse. The oscillations of the voltage and current waveforms are from the
switching of micro-pulses.

The effect of charging voltage from 350 to 450 V is then studied. A same recipe using 10-8 micro-pulses and 5 mTorr are used. According to Figure 7.2, Dramatic increase of the pulse current from 20 to 300 A can be seen. Unfortunately, the charging voltage can’t be set to too high values due to the arcing issue.

![Figure 7.2 I-V curves of the MPP discharge under different charging voltages. Recipe of $\tau_{off} = 10$ µs, $\tau_{on} = 8$ µs and 5 mTorr are used.](image)

The effect of pressure is shown in Figure 7.3. The 10-8 recipe and 450 V charging voltage are used. Voltage can be seen to drop considerably as the pressure increases from 1 to 15 mTorr. The current almost doubles. This is trend is commonly seen in plasma discharge.

As comparing with HiPIMS discharge, the peak voltage and current in MPP are lower. In the HiPIMS operation peak voltage and current are typically 900 V and 750 A. However, the high current stage in MPP can last for a much longer time than in HiPIMS discharge (1000 µs vs. 100 µs).
Figure 7.3 I-V curves of the MPP discharge under different pressures. $\tau_{\text{off}} = 10 \, \mu s$, $\tau_{\text{on}} = 8 \, \mu s$, 450 V.

7.1.2 Triple Langmuir Probe Diagnostics

Triple Langmuir probe measurements of the MPP plasma are then performed. Figure 7.4 shows the measured $T_e$ and $n_e$ for recipes with different $\tau_{\text{on}}$. The temporal evolution of $n_e$ basically resembles the corresponding current curve. At the initial 1500 $\mu$s when weak pulses (40-6) are used, $n_e$ is quite low. Higher density is achieved when main pulses are used. By increasing the $\tau_{\text{on}}$ from 6 to 9 $\mu$s, the electron density near the substrate increases from $2 \times 10^{17}$ to $8 \times 10^{17}$ m$^{-3}$. Meanwhile, $T_e$ slightly decreases but is basically within the range of 2 to 4 eV after the plasma being ignited. As compared with the HiPIMS discharge, the density in MPP discharge is comparable but $T_e$ is lower. Electrons in HiPIMS may not thermalize effectively due to the short pulse length. In MPP discharge, the behaviors after the pulse ends are not as important as in HiPIMS. No obvious second peak is observed.

By keeping $\tau_{\text{on}}$ the same at 10 $\mu$s, $\tau_{\text{off}}$ is varied from 8 to 16 $\mu$s. Correspondingly $T_e$ increases and $n_e$ decreases (Figure 7.5). The shorter $\tau_{\text{off}}$ implies less loss of plasma to build up a denser plasma. More Coulomb collisions in a higher density plasma result in a lower $T_e$.  

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Figure 7.4 Triple Langmuir probe measurements of (right) \(n_e\) and (left) \(T_e\) in a MPP sputtering plasma. Pulse off-time is 10 µs, while the on-time varies from 6 to 9 µs.

Figure 7.5 Effect of micropulse off-time (right) \(n_e\) and (left) \(T_e\) in MPP. \(\tau_{off} = 8-16\) µs, \(\tau_{on} = 10\) µs, 450 V, 5 mTorr.

The effects of \(V_{ch}\) and pressure on the plasma are measured in Figure 7.6 and Figure 7.7 respectively. A higher charging voltage produces higher \(n_e\) but slightly lower \(T_e\). A higher pressure basically increases the \(n_e\) and reduced the \(T_e\).
7.1.3 Deposition Flux

The Al ionization fractions in the deposition flux are measured on the substrate level. Figure 7.8 shows the results with different micropulse on-time, repetition rate, and pressure. Micropulses of 10 µs off and 6 µs on produces very low ionization, basically not detectable by our GEA/QCM tool. A longer pulse on-time gradually increases the ionization fraction to about 13%. Increasing the repetition frequency doesn’t affect the ionization fraction. The pressure also influences the ionization process, basically decreasing the fraction when it is reduced. Through the whole test, the ionization fraction of Al in a MPP discharges are below 15% on the substrate, which is much lower than that in the HiPIMS discharge (~29%). The reason may lie in the facts that HiPIMS discharge has a much higher pulse voltage and thus higher electron energy to effectively ionize atoms. Also MPP discharge
uses very short pulse off-time so that ions may not be efficiently released from the target region.

Figure 7.8 Ionization fraction of Al during MPP discharge when using different (a) micropulse on-time, (b) repetition rate, and (c) pressure.

The deposition rates are measured by increasing the pulse on-time. In order to compare with the HiPIMS discharge, the deposition rates are plotted against the average power. About 40 nm/min at an average power of 2 kW and over 50 nm/min at 4 kW are achieved (Figure 7.9). That is almost twice higher than that in HiPIMS. Obviously, the higher deposition rates in MPP discharge are advantages over the HiPIMS discharge.

Figure 7.9 Deposition rates measured versus average power in MPP discharge. The increase of average power was achieved by increasing the micropulse on-time.
7.2 MPP Cyprium

A different model of the Zpulser MPP generator has also been studied. In this Cyprium model, the pulsing voltage can very rapidly drop during the off-time. The concept is that if the voltage can drop to zero, the extraction of ions generated during the pulse on-time is more efficient. The pulsing file is still about the same with micropulse on- and off-time being the control parameters.

7.2.1 Discharge I-V Characteristics

Using this Cyprium model, the effects of micropulse on-time on the I-V characteristics are first studied, as shown in Figure 7.10. Each pulse is about 1000 µs long. Here 10-8 in the figure means 10 µs off and 8 µs on. The charging voltage is 350 V and pressure is 5 mTorr. A first glance at the waveforms one can see a lot of oscillations for both voltage and current. And this is actually the distinctive feature for Cyprium model. The voltage, however, does not reduce all the way to zero. A longer $\tau_{\text{on}}$ leads to a higher current, which can be seen more clearly in the blowup between 500 and 600 µs. Both the voltage and current waveforms have been distorted.

A shorter off-time generates higher current according to Figure 7.11, so does higher charging voltage according to Figure 7.12.

Figure 7.10 I-V curves of the MPP Cyprium under different $\tau_{\text{on}}, \tau_{\text{off}} = 10\ \mu\text{s}, \tau_{\text{on}} = 8-12\ \mu\text{s}, 350\ \text{V}, 5\text{mTorr}$. The right figure shows the blow-up of the I-V curves.
7.2.2 Triple Langmuir Probe Diagnostics

The effects of micropulse on-time, off-time and charging voltage on the time-resolved $T_e$ and $n_e$ are investigated and shown in Figure 7.13 to Figure 7.15. Due to the large amplitude of oscillation, curves for different conditions can be overlapped. Basically, a longer pulse on-time generates higher $n_e$ which is consistent with the discharge current measurements. A longer off-time usually leads to a lower $n_e$ and about same $T_e$. A higher charging voltage obviously increases the plasma density and reduces the $T_e$.
Figure 7.13 Effect of $\tau_{on}$ on $n_e$ and $T_e$ in MPP Cyprium. $\tau_{off}$ 10 $\mu$s, $\tau_{on}$ 8-12 $\mu$s, Vch 350 V, 5 mTorr.

Figure 7.14 Effect of $\tau_{off}$ on $n_e$ and $T_e$ in MPP Cyprium. $\tau_{on}$ 10 $\mu$s, $\tau_{off}$ 8-16 $\mu$s, Vch 350 V, 5 mTorr.

Figure 7.15 Effect of charging voltage on $n_e$ and $T_e$ in MPP Cyprium. $\tau_{off}$ 10 $\mu$s, $\tau_{on}$ 8 $\mu$s, Vch 300-400 V, 5 mTorr.
Figure 7.16 3D measurement of $T_e$ and $n_e$ during the MPP Cyprium discharge.

Figure 7.17 3D distribution of $n_e$ in MPP Cyprium discharge.
3-D TLP measurements can also be performed for MPP plasma characterization. Figure 7.16 shows the plasma from 0 to 14 cm in radial direction and from z = 13 cm to z = 3 cm moving toward the target. The recipe is $\tau_{\text{off}}$ 10 µs, $\tau_{\text{on}}$ 8 µs, $V_{\text{ch}}$ 350 V, and 5 mTorr. $n_e$ is seen to be slightly higher at $r=12$ cm on the substrate level, and increase dramatically when getting closer to the target. In order to eliminate the effect caused by waveform oscillation, the maximum values of the $n_e$ between 500 and 600 µs are used to plot the $n_e$ distribution in the chamber, as shown in Figure 7.17. This is actually similar to the expansion peak distribution in HiPIMS discharge. The plasma near the race track is very dense and non-uniform. Moving away from the target, $n_e$ decreases while its profile becomes more and more uniform.

Similar method can be used to present the dependence of $n_e$ at different location on the pulsing parameters. As shown in Figure 7.18, $n_e$ increases with longer on-time, shorter off-time and higher charging voltage. The trend is valid for various locations at different z.

![Figure 7.18](image)

Figure 7.18 Effect of pulsing parameters on $n_e$ at different z locations. (a) Different $\tau_{\text{on}}$, (b) different $\tau_{\text{off}}$, and (c) different $V_{\text{ch}}$. 

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MPP Cyprium is then operated in different magnetic configurations of 200, 500, and 800G. A recipe of 10-10, 350V, 5mTorr is used. The IV curves are compared in Figure 7.19. With an increased B field strength, discharge voltage decreases and current increases. This is expected with a better magnetic confinement to enhance the plasma.

![IV curves of MPP Cyrium discharge in different magnetic configurations of 200, 500, and 800G.](image)

The T_e, n_e and V_f curves are then compared at two different locations, z = 13 cm near the substrate and z = 3 cm near the target, in Figure 7.20 and Figure 7.21 respectively. Radial position is fixed at r = 12 cm. At z = 13 cm, T_e is about the same around 2 eV in 500 and 800G, slightly lower in 200G. n_e basically increases with higher B field, from 5×10^{17} m^{-3} to about 1.5×10^{18} m^{-3}. The floating potential becomes more negative when changing from 200 to 800G. At 800G, V_f can be as low as -10 V and doesn’t vary very much during the pulse. The floating potential is comparable to the HiPIMS case.

At z = 3 cm, T_e is about the same for all three configurations, at about 1.8 eV in average. n_e increases more dramatically with higher B field, from 5×10^{18} m^{-3} to about 2.5×10^{19} m^{-3}. The floating potential becomes more negative, oscillating between 0 and -10 V for 200G, -10 to -30 V for 500G and -20 to -45 V for 800G.
Figure 7.20 Comparison of $T_e$, $n_e$, and $V_f$ at $r=12$ cm and $z = 13$ cm in three configurations.

Figure 7.21 Comparison of $T_e$, $n_e$, and $V_f$ at $r=12$ cm and $z = 3$ cm in three configurations of different magnetic field strength.
7.2.3 Ionization Fraction

The Cu ionization fractions in the deposition flux are measured on the substrate level in all three configurations of 200, 500, and 800G. Figure 7.22 showed the results with different charging voltage, micropulse on- and off-time, and pressure. The baseline recipe is 10-8 pulses, 350 V, 5 mTorr in 500G. Figure (a) shows that by increasing the charging voltage, Cu⁺ fraction increases up to 30%, which is less than that in HIPIMS but higher than in DC. Fig. 20(b) shows the effect of pulse on-time. In 200G configuration, Cu⁺ fraction increases with a longer on-time, while in 800G it is overall much lower and follows an opposite trend. In Fig. 20 (c), it shows that in all three magnetron configurations, Cu⁺ fraction increases with a longer pulse off-time, especially obvious for the 800G case. In Figure 7.22 (d), it shows Cu⁺ fraction increases with a higher pressure from 15 to 35%.

The effect of charging voltage lies in the fact that higher charging voltage discharges generated higher-density plasma according to the triple probe measurements. Longer $\tau_{on}$ also increases $n_e$ while and $T_e$ remains about the same, so that higher ionization fraction is achieved. The exceptions for 14µs pulses and 800G B field configuration are probably because the very high $n_e$ produced a plasma potential gradient to prevent efficient ion extraction. Such an effect by varied B field has been seen in the previous HIPIMS experiments too. As for the effect of pulse off-time $\tau_{off}$, it is believed that a long enough off-time is required for plasma (ions) to diffuse to substrate. According to the experiments using HIPIMS, it takes more than 100 µs to receive the expanding plasma flux on the substrate. The enhancement of Cu⁺ fraction in the deposition flux by long $\tau_{off}$ is especially important for 800G B field configuration, because of its already low ion extraction efficiency. The higher
pressure means more frequent electron-atoms collisions for more ionizations, which is evidenced by higher $n_e$ measured.

Figure 7.22 Ionization fraction of Cu in the deposition flux vs. varied (a) charging voltage, (b) micropulse on-time, (c) micropulse off-time, and (d) pressure. The baseline recipe is 10-8 pulses, 350 V, 5 mTorr in 500G. Different B field strengths were used.
7.3 Conclusions

MPP is a relatively new technique as compared with HiPIMS. Our group of CPMI is likely one of the few to have two different models of the MPP plasma generators. The study here is believed to greatly contribute to the understanding of this unique pulsing technique. Similar to what we did for HiPIMS, MPP discharge is first systematically characterized on a planar magnetron before being applied for trench metallization tests. The unique features of the MPP discharge originate from its flexibility in adjusting the arrangement of micro-pulses. Custom pulse shapes can be achieved. The I-V characteristics show multiple stages including one with high discharge currents. The Cyprium model is even able to strongly oscillate the voltage and current pulses.

Similar plasma densities during the pulses are achieved in MPP as in HiPIMS, though there is no obvious high-density expansion peak. Stronger plasmas can be achieved using longer micro-pulse on-time, shorter off-time and higher charging voltage. The deposition rates are about twice higher than those in HiPIMS at the same average power. The ionization fractions are shown to increase with longer micro-pulse on-time, higher charging voltage, etc. It also has a similar dependence on the B field strength. Overall the ionization fractions by MPP are much lower as compared with HiPIMS. This is likely because of the too short off-time (on the order of 10µs) prevents the ion release, even for the Cyprium model with designs of reducing voltage between micro-pulses.

In all, MPP and HiPIMS have some their own advantages and disadvantages. For MPP, it is favored for its less loss of deposition rate. Since it has much longer pulse duration, the plasma potential in the bulk plasma region is quite flat, being close to that in DC. As a result, the ions are more easily extracted to the substrate (though it still has lower ionization fraction on substrate than in HiPIMS). To a certain extent, MPP is an intermediate technique between DC and HiPIMS. It uses longer discharge duration while maintaining the short pulses, although these short pulses are not as powerful as the individual pulses in HiPIMS. As a result, almost all the plasma parameters of MPP discharge is in between those of DC and HiPIMS. For industrial applications, choice between MPP and HiPIMS should be based on the specific plasma requirements. For barrier/seed deposition, high enough ionization fraction is a more important parameter than deposition rate, especially with the film deposition only lasts for 10 seconds or so.
Chapter 8  Study on Hollow Cathode Magnetron

Hollow cathode magnetron is featured with high ionization fraction by one single source. The concept is to combine this advantage with the good ionization capability by HiPIMS or MPP to achieve more highly ionized plasma to improve the metallization in trenches or vias. To test the concept, plasma diagnostics are performed to compare the DC sputtering of Cu as the base line and the pulsed sputtering. Ionization fraction is measured and used as an important metric to develop the deposition recipes. The promising recipes are subsequently applied for Cu deposition on patterned silicon wafers with narrow trench structures. The goal is to obtain conformal deposition. A scanning electron microscope (SEM) is used to yield high-quality magnified images of patterned silicon wafers under different deposition conditions for comparison.

8.1  Plasma Studies

8.1.1  DC Operation in Hollow Cathode Magnetron

DC magnetron sputtering (DCMS) in the hollow cathode magnetron is first explored. Key parameters such as the electron density and temperature, their spatial distribution, ionization fraction and deposition rate are characterized.

Using a single Langmuir probe, \( n_e \) and \( T_e \) are measured. Error! Reference source not found. shows the dependence of \( T_e \) and \( n_e \) on the input power and pressure. The pressure is varied from 1 to 10 mTorr in Ar and the power is increased from 4 to 16 kW. The measurements are done at 1 inch above the substrate. A higher input power produces both higher \( n_e \) and \( T_e \). Such a trend is especially prominent at higher pressure. At 10 mTorr, \( n_e \) dramatically increases from \( 8 \times 10^{16} \) cm\(^{-3} \) to \( 5 \times 10^{17} \) cm\(^{-3} \) while \( T_e \) increases from 1.9 to 3.0 eV. At lower pressure, the advantage of higher power is less obvious. At 1 and 2 mTorr, \( n_e \) and \( T_e \) are comparable. The effect of pressure is easy to see. A higher pressure leads to a larger \( n_e \) and a lower \( T_e \). In order to establish a strong plasma, it is important to have high power input as well as to effectively induce the power into the plasma and heat the electrons. The latter usually requires more frequent electron-neutral collisions, for example, by using higher pressure. At lower pressure, there are fewer atoms as the “fuel” to be ionized. As a result, the ionizations are fewer, the electron density is lower, and the electron temperature is higher due to less
energy loss from collisions. The effect of power input is suppressed because this low efficiency for the power assumption by plasma. At a higher pressure, however, more electron-atom collisions lead to a higher electron density and reduced electron energies.

Figure 8.1 T\textsubscript{e} and n\textsubscript{e} of Ar sputtering plasma at different pressures (1-10 mTorr) and power (4 and 16 kW), 1 inch above the substrate).

To have an idea of the plasma distribution in the substrate region, probe was scanned from near the substrate to a height of 5 inch (12.7 cm) and from the center to the 10 cm away. The results are shown in Figure 8.2 and Figure 8.3 respectively. The plasma is observed to be denser and hotter at a higher position, i.e., closer to the target. At a same height, plasma is seen to be non-uniform, with a higher n\textsubscript{e} and T\textsubscript{e} at the center. The observations are consistent with the fact that plasma diffuses from the opening of the hollow cathode to the substrate and the side wall. It should be mentioned that the uniformity of the plasma can be improved in HCM by adjusting the downstream magnetic field using two EM coils, though it’s not the main focus of my study and does not have a foreseeable effect on the proposed tests.
The ionizing capability in HCM by DC sputtering is determined through Cu ionization measurements. The same setup of GEA/QCM assembly is installed at the center of the pedestal. Varied power and pressure in their typical ranges are studied (Figure 8.5). At 5 mTorr, the Cu ionization fraction in the deposition flux increases from a few percent to above 20% at 20 kW. The increasing trend gradually slows down at higher power. At 2 mTorr, Cu IF shows a similar dependence on the power but the overall values are lower. Only slightly over 10% is achieved. This is believed to be from the lower \( n_e \) in the plasma at a lower pressure.
Figure 8.4 Ionization fraction of Cu in DC hollow cathode magnetron sputtering.

Figure 8.5 Deposition rate of DC HCM sputtering as a function of power and pressure, at both the center and edge position.

Deposition rates by DC sputtering are measured at both the center and edge of the pedestal using QCM. As shown in Figure 8.5 (a), the deposition rate increases proportionally with the input power and is about 16 Å/s at 10 kW. There is certain non-uniformity across the wafer. The deposition rate at center is slightly higher than that at edge. The deposition rate decreases with an increased pressure, as shown in Figure 8.5 (b). At higher pressures, atoms undergo more collisions so a larger fraction are scattered towards the chamber wall before reaching the substrate.
### 8.1.2 MPP (Solo) in Hollow Cathode Magnetron

In this section, MPP discharge in the INOVA HCM is studied. Table 8.1 shows the performed tests.

| Table 8.1 Tests of MPP sputtering done in INOVA hollow cathode magnetron |
|-------------------------------------------------|-----------------|-----------------|-----------------|---------|------------------|
| Pulising parameters                             | Pressure         | Various locations |
| Charging voltage                               | Micropulse on- and off-time | Macropulse length | Repetition frequency/ power |
| I-V characteristics                             | ×                | ×                | ×                | ×       | ×                |
| Triple probe                                   | ×                | ×                | ×                | ×       | ×                |
| Ionization fraction (metal)                     | ×                | ×                | ×                |         | ×                |
| Deposition rate                                 | ×                | ×                | ×                |         |                  |

The I-V discharge characteristics of the MPP generator were studied prior to the triple probe measurements. The typical I-V curves of the MPP discharge show multiple stages, as seen in Figure 7.1. The macro-pulse is programmed to be 3000 μs long, consisting of micro-pulses of $\tau_{\text{off}}$ 40 μs and $\tau_{\text{on}}$ 6 μs (or shorted as 40-6) in the initial 1500 μs and a session of micro-pulses to gradually ramp up. The weak recipes at the beginning make sure the discharge can be ignited. The main part of the macro-pulse is the last 1000 μs, consisting of the micro-pulses of $\tau_{\text{off}}$ 10 μs and $\tau_{\text{on}}$ 6 to 9 μs. The charging voltage is 450 V and pressure is 5 mTorr. Plasma ignition occurs in the first 100 μs ending with a small surge of current. Then it goes into a low power sputtering mode similar to the conventional DC magnetron sputtering. After the session of higher-duty-ratio micro-pulses starts, the current is dramatically ramped up and may enter the steady state given enough time. In this last stage, the current is very high to effectively ionize the sputtered material.

The same or similar MPP pulse recipes used in planar magnetron tests are used here. The pulse files usually include a session of weak micropulses and then ramp up to the main micropulses with desired $\tau_{\text{off}}$ and $\tau_{\text{on}}$. Other controlling parameters including the charging voltage $V_{\text{ch}}$, repetition frequency $f$ and pressure are also varied. The ranges of the pulsing and discharge parameters are limited by difficulty to ignite and maintain a stable discharge.
Similar voltage and current waveforms are obtained in HCM as those in planar magnetron. Figure 8.6 shows one example of typical voltage, current and power waveforms. More than 500 V as the peak voltage and almost 350 A as the peak current are achieved using 10-16 ($\tau_{\text{off}} = 10 \mu s$, $\tau_{\text{on}} = 16 \mu s$) main pulses. The corresponding peak power reaches 180 kW.

The peak discharge voltage and current are measured for different pulsing recipes. Figure 8.7 shows an example for different $V_{\text{ch}}$ and $\tau_{\text{on}}$. Higher $V_{\text{ch}}$ is favored, which increases the discharge voltage and current for each pulse profile. A longer $\tau_{\text{on}}$ also produces high peak voltage and peak current. More tests (not shown here) reveal that a longer $\tau_{\text{off}}$ reduces the discharge voltage and current. The repetition frequency does not obviously affect the discharge. A higher discharge voltage and a lower current are obtained at a lower pressure.

![Figure 8.6 Typical I, V waveforms using MPP Solo plasma generator in HCM.](image-url)
Figure 8.7 Discharges using different pulse profiles ($\tau_{\text{off}} = 10$ µs, and $\tau_{\text{on}} = 8$ to 16 µs) with different charging voltages.

Triple Langmuir probe measurements of the $T_e$ and $n_e$ are performed at 1 inch above the pedestal. Results for varied $V_{\text{ch}}$, $\tau_{\text{on}}$ and pressure are shown here. By increasing $V_{\text{ch}}$ to 500 V, the electron density increases up to $3.5\times10^{18}$ m$^{-3}$ (Figure 8.8). The pulse profile is 10-14 and the average power is kept at 2 kW. Meanwhile, $T_e$ slightly decreases from about 4 to 2 eV. A decrease of $T_e$ when the plasma intensifies is commonly seen. As a comparison, DC magnetron sputtering only generates a density of about $1\times10^{17}$ m$^{-3}$ even with the power increased to 16 kW or $5\times10^{17}$ m$^{-3}$ with a higher pressure of 10 mTorr. A great enhancement of plasma density is achieved.

Figure 8.8 $T_e$ and $n_e$ of the plasma generated by MPP sputtering of Cu in HCM using different $V_{\text{ch}}$. Recipes are $\tau_{\text{off}}$ 10 µs, $\tau_{\text{on}}$ 14 µs, $V_{\text{ch}}$ 350-450 V, 5 mTorr.

Then $\tau_{\text{on}}$ is increased from 8 to 18 µs, with $\tau_{\text{off}}$ set at 10 µs and $V_{\text{ch}}$ at 450 V. The corresponding plasma measurements are shown in Figure 8.9. The density increases to $3.5\times10^{18}$ m$^{-3}$ while $T_e$ drops to
about 2 eV. If comparing with the planar magnetron discharge, similar peak density is achieved on HCM when a same recipe is used. But the HCM allows usage of more aggressive pulse profiles, e.g. longer $\tau_{\text{on}}$. As a result, an easy-to-achieve density on HCM is higher than that in planar magnetron.

Figure 8.9 $T_e$ and $n_e$ for different $\tau_{\text{on}}$ on-time from 8 to 18 µs. Recipes are $\tau_{\text{off}}$ 10 µs, $\tau_{\text{on}}$ 8 to 18 µs, $V_{\text{ch}}$ 450 V, 5 mTorr.

The effect of the pressure on $T_e$ and $n_e$ are shown in Figure 8.10. It can be seen that the pressure presents a familiar effect on the plasma properties. Lower pressures leads to a much lower $n_e$ but slightly higher $T_e$. This is due to the same reason mentioned previously that there are fewer electron-atom impact ionizations. Overall $n_e$ is quite high during the pulse, which is beneficial for high ionization. The electron temperature is comparable to the DC case, presenting no additional harm to the substrate deposition.

Figure 8.10 $T_e$ and $n_e$ for the discharge at different chamber pressure. Recipes are $\tau_{\text{off}}$ 10 µs, $\tau_{\text{on}}$ 14 µs, $V_{\text{ch}}$ 450 V, 1-5 mTorr.
After confirming high $n_e$ can be achieved during pulses, Cu ionization fractions (IF) on the substrate level are measured. Pulses with different micropulse on-time $\tau_{on}$ are chosen and are used at different average power, pressure and $V_{ch}$. Figure 8.11 (a) shows the results of Cu IFs with different $\tau_{on}$ from 8 to 16 µs. Different average power is obtained by adjusting the repetition frequency automatically by the generator. The effect of longer $\tau_{on}$ is obvious that the ionization fraction is increased to about 25% using a $\tau_{on}$ of 16 µs. This is easy to understand considering denser plasmas being generated with longer $\tau_{on}$ as observed in Figure 8.9. Increasing the average power doesn’t increase the ionization fraction. The repetition frequency does not affect the behavior within each separate macropulses. As a comparison, the Cu IFs in DC magnetron sputtering are shown in the figure. At 2 and 4 kW, there is only less than 10% of Cu being ionized, substantially lower than that in the MPP Solo operation. However, it should be mentioned that the ionization fraction in DCMS does increase with a higher power and is capable of increases to above 20% as the power goes up to 20 kW, which becomes comparable with the 25% in MPP sputtering.

![Figure 8.11 Ionization fraction of Cu during MPP Solo sputtering in HCM. Different micropulse on-time from 8 to 16 µs are used while off-time was 10 µs. (a) $V_{ch}$ 400V, 5 mTorr, (b) $V_{ch}$ 400V, 2 mTorr, and (c) varied $V_{ch}$, 5 mTorr.](image)

Figure 8.11 (b) shows the results for different pressure. The ionizations are generally lower at 2 mTorr than at 5 mTorr, which is expected. The increase of IF with higher $\tau_{on}$ can also be observed. The decreasing trend with higher average power may be because too high frequency has to be used to
affect the effective capacitor charging.

Figure 8.11 (c) shows the effect of the charging voltage. For either pulse file, a higher $V_{ch}$ leads to an increased Cu IF. Unfortunately, too high $V_{ch}$ usually causes unstable discharge, even with an average power well below 10 kW allowed by the power supply.

The deposition rates during MPP Solo sputtering are also measured and compared with the DCMS. Different $\tau_{on}$ is used while $V_{ch}$ is fixed at 450 V. In order to compare with DCMS, the data by MPP sputtering are also plotted against the average power, as shown in Figure 8.12. The deposition rates by MPP sputtering are similar to those by the DCMS or slightly lower as the average power increases. This is a possible advantage of MPP over typical HiPIMS which usually comes with some significant deposition loss at a same average power. In both DC and MPP sputtering, a good uniformity of deposition rates on the wafer level can be seen.

![Figure 8.12 Comparison of deposition rates in DC and MPP magnetron sputtering.](image)
8.1.3 MPP (Cyprium) in Hollow Cathode Magnetron

Characterizations of the discharge using MPP Cyprium model in HCM are described in this section. The discharge IV characteristics, the time resolved $T_e$ and $n_e$ behaviors and the Cu IF on the substrate level are measured. The pulse profiles used for the study are the same as the ones used for planar magnetron study.

Figure 8.13 Voltage and current waveforms using MPP Cyrium plasma generator in HCM, (a) vs. $\tau_{on}$ with recipes of $\tau_{off}$ 10 $\mu$s, $\tau_{on}$ 8-16 $\mu$s, $V_{ch}$ 350 V, 5 mTorr, (b) vs. $V_{ch}$ with recipes of $\tau_{off}$ 6 $\mu$s, $\tau_{on}$ 10 $\mu$s, $V_{ch}$ 300-375 V, 5 mTorr, and (c) vs. pressure, with recipes of $\tau_{off}$ 10 $\mu$s, $\tau_{on}$ 10 $\mu$s, $V_{ch}$ 350 V, 2-10 mTorr.
The voltage and current waveforms at different conditions are measured. Figure 8.13 (a) shows a set of recipes with different $\tau_{on}$ from 8 to 16 $\mu$s. The rest parameters are $\tau_{off}$ 10 µs, $V_{ch}$ 350 V, repetition frequency 30 Hz, pressure 5 mTorr. The oscillations of voltage and current due to the micropulses can be seen. The voltage basically oscillates between -200 and -600 V, and does not decrease to zero. A higher current up to 600 A can be achieved by using longer $\tau_{on}$. This is higher than the typical pulse current by MPP Solo model.

The current can also be increased by using higher $V_{ch}$ (Figure 8.13 b). The pulse profile used here is 6-10. High peak current up to 600 A can also be obtained. However, it is difficult to sustain this high current through the entire pulse. Figure 8.13 (c) shows the effect of different pressures from 2 to 10 mTorr. A recipe profile 10-10 is used. A much higher discharge current is seen at 10 mTorr, while the pulse voltage becomes lower.

The triple Langmuir probe measurements are then performed at 1 inch above the substrate. All the above three sets of recipes are tested. The results are shown in Figure 8.14. Figure (a) shows that by increasing $\tau_{on}$, the electron density is greatly increased up to $5 \times 10^{18}$ m$^{-3}$. This is almost two orders of magnitude higher than that in DCMS, and is also higher than the MPP Solo operation. This high $n_e$ doesn’t last the entire pulse duration though and starts to drop after about 500 $\mu$s. $T_e$ is following opposite trends that it decreases with longer $\tau_{on}$. Over time it decreases to a minimum of about 2 eV at about 500 $\mu$s and then goes up. The overall higher $n_e$ is believed to relate to a different pulsing mechanism as in MPP Solo model. In Cyprium model, the discharging and charging of the capacitors in the pulsing unit are faster so the discharge voltage drops significantly and increases to a high value after each micropulse. Instead MPP Solo model keeps about a constant discharge voltage during the macropulse. When the discharge voltage drops, it helps the release of confined plasma to the substrate region. Then the voltage oscillates to a much higher value than the almost steady-state voltage during MPP Solo discharge. This provides electrons with higher energy for ionizations. In another word, the oscillating voltage allows a more efficient power transfer to the plasma. The decrease of the $n_e$ in the later part of the macropulse is likely because the density inside the hollow cathode has become so high that the electrons are considerably thermalized to have very low energies and lose the capability of efficient ionization. Because of this reason, using longer macropulse won’t really improve the performance.
Figure 8.14 (b) and (c) then show the $T_e$ and $n_e$ at different $V_{ch}$ and pressure. By increasing the charging voltage, very dense plasma (even on the substrate level) is achieved (up to $8\times10^{18}$ m$^{-3}$ at 375 V). $T_e$, on the other hand, decreases to as low as 1 eV. Similar behaviors of reducing $n_e$ in the latter half of the macropulse are observed. A higher pressure produces a plasma with higher $n_e$ and lower $T_e$ as expected.

Figure 8.14 $T_e$ and $n_e$ for different (a) $\tau_{on}$, (b) $V_{ch}$ and (c) pressure using MPP Cyrium plasma generator in HCM. The same recipes in the above IV characteristic tests (Figure 8.13) are used correspondingly.
Similar Cu ionization fractions (IF) as in MPP Solo operation are done on the substrate level. The influences of micropulse on-time $\tau_{on}$, $V_{ch}$ and pressure are first studied. Figure 8.15 shows the results of Cu IFs with different $\tau_{on}$ from 8 to 16 µs, different $V_{ch}$ of 300 and 350 V, and different pressure of 2 and 5 mTorr. By increasing $\tau_{on}$, Cu IF basically increases, for example from about 15 to 22% at a $V_{ch}$ of 350 V and 5 mTorr. This is consistent with the high plasma density obtained when using longer $\tau_{on}$. A higher $V_{ch}$ leads to an increased Cu IF by approximately 40%. Cu IF also scales with a higher pressure.

![Graph showing Cu IF vs. Pulse on time](image)

**Figure 8.15** Ionization fraction of Cu during MPP Cyrium sputtering in HCM. Different micropulse on-time from 6 to 16 µs are used while off-time is 10 µs. $V_{ch}$ is varied from 300 to 400 V. Pressure is varied from 2 to 5 mTorr.
8.1.4 HiPIMS Operation

Finally, the HiPIMS discharge on HCM using Huettinger plasma generator is characterized. The pulsing parameter $V_{ch}$ is varied between 700 and 1200 V, and pulse duration $t_p$ of 50 and 100 µs are used.

![I-V characteristics of HiPIMS on HCM at different charging voltage, (a) 700-1200 V for 50 µs pulses, and (b) 700-1000 V for 100 µs. The other parameters are repetition frequency of 100 Hz and 5 mTorr in Ar. Note the scales are different.](image)

The I-V characteristics of the HiPIMS on HCM are measured. Figure 8.16 (a) and (b) show the results for 50 and 100 µs pulses respectively. Basically they have similar waveforms as observed in the plasma magnetron sputtering, with a dramatic increase of pulse current during the pulse. Higher $V_{ch}$ and longer pulse duration result in higher discharge current. Some differences can be seen though. HCM allows high charging voltages with very stable discharge. Also the discharge voltages of higher values are maintained in HCM, while on planar magnetron they quickly reduce to about 400 V for
typical the recipes. The pulse current does not show sign of saturation even in the 100 µs pulses. The reason for such differences is believed to be the better electron confinement in HCM. In planar magnetron, too high discharge voltage leads to high energy electrons to escape the magnetic confinement. High enough current leads to the high-density plasma to very effectively diffuse out of the race track region as well. Such plasma loss mechanisms, on the other hand, are greatly suppressed in HCM.

![Figure 8.17 T_e and n_e of HiPIMS plasma on HCM at different charging voltage, (a) 700-1200 V for 50 µs pulses, and (b) 700-1000 V for 100 µs. The other parameters are repetition frequency of 100 Hz and 5 mTorr in Ar.](image)

Triple Langmuir probe measurements are performed for these two sets of recipes. The time resolved T_e and n_e are plotted in Figure 8.17. Again, they also have similar behaviors as those in the planar magnetron. For 50 µs, T_e decreases during the pulse to about 3 or 4 eV, while n_e increases.
After the pulse ends, plasma expansion out of the hollow cathode leads to more peak structures. A higher charging voltage produces a peak electron density as high as about $2 \times 10^{18}$ m$^{-3}$. By using longer pulses (100 µs), higher electron densities are achieved both during the pulse and after the pulse in the expansion plasma.

Cu ionization fractions (IF) on the substrate level are measured under different pulse parameters. Figure 8.18 shows the results of Cu IFs with different charging voltages. Three different pulse durations, 50 to 150 µs, are used. Cu IF basically increases with the charging voltage. For 50 µs pulses, it increases from 10 to 19%. For 100 µs, it increases from 18 to 29%. By increasing the pulse duration, Cu IF also increases from 10 to 20% for the recipes of 1000 V charging voltage. It is noted that using a same recipe, the Cu IF in HCM is lower than that in planar magnetron, even its 800 Gauss configuration. This is likely another effect of the very strong electron confinement. The study in planar magnetron shows a stronger magnetic field reduces the ionization fraction in deposition flux due to the inefficient ion extraction.

Figure 8.18 Ionization fraction of Cu during MPP Cyrium sputtering in HCM. Different micropulse on-time from 6 to 16 µs are used while off-time is 10 µs. $V_{ch}$ is varied from 300 to 400 V. Pressure is varied from 2 to 5 mTorr.
8.2 Trench Deposition

With all the plasma studies conducted, we have achieved a better understanding of the HiPIMS and the MPP discharge. Meanwhile, we have developed a wide range of promising recipes. The performance of HiPIMS and MPP magnetron sputtering for interconnect metallization will be further tested using these recipes in the 200mm HCM as compared with DCMS. Cu is deposited on patterned wafers with trenches of different widths as narrow as 70 nm. The conformality of the Cu film coverage on the trench will be compared using scanning electron microscopy (SEM).

8.2.1 Trench Deposition using DCMS

Trench deposition using DC magnetron sputtering is first performed as the baseline. The used patterned trench structures can be seen in Figure 8.19. To better reveal the weak point and evaluate the performance, narrowest trenches available are used, which are the 70 nm and 80 nm ones as shown. These trenches show quite clear and wide openings at the top before deposition. About 350 Å thick film is then deposited on the wafer. Depending on the processes, different degree of overhang will show up and is characterized using SEM for the process evaluation. Among all the potential weak points during interconnect metallization such as poor step coverage or weak bonding between barrier and seed, the overhang built up at the top is the easiest and most accurate to measure and thus adopted in this work as the criterion for comparison.

![Patterned wafer with 70 nm (left) and 80 nm (right) trench structures before deposition.](image)

For DCMS, two different power inputs of 4 and 16 kW at both 2 and 5 mTorr are used for trench deposition. The trenches after deposition are measured using SEM. Figure 8.20 and Figure 8.21
show the images for 5 mTorr and 2 mTorr respectively. The deposited films can be clearly seen on top of the trenches. The openings at the top become narrower due to overhang formation. This is more obvious for the 70 nm trenches than for the 80 nm ones.

![Figure 8.20 SEMs of the 70 and 80 nm trenches after Cu deposition by DCMS at 5 mTorr.](image)

![Figure 8.21 SEMs of the 70 and 80 nm trenches after Cu deposition by DCMS at 2 mTorr.](image)

Although it can be roughly determined that the trench opening is larger at higher power, it is difficult to make any quantified comparison. For this purpose, a special procedure is used to measure the overhang size by analyzing the cross-section SEMs. As illustrated in Figure 8.22, the widths of the space between trenches are measured at the widest ($w_1$) and narrowest point ($w_2$). The value of $(w_1 - w_2)/2$ is used to define the excessive material building up at the top. It is then divided by the field deposition thickness $d$, to account for the effect of enhanced overhang formation by thicker film deposition. This ratio is defined as the overhang/dep ratio or shorted for overhang ratio. The lower this
value, the more conformal is the film coverage on the trenches. Measurements are conducted on different trenches and averaged. The standard deviation is contributed by two factors. One is the uncertainty to decide the widths from the SEMs, and the other is the line edge roughness of the trenches.

![SEM image of trenches](image)

*Figure 8.22 Method to analyze the overhang using cross-section SEMs of trenches.*

The overhang/dep ratios for the DCMS recipes are plotted in Figure 8.23. At 5 mTorr, 16 kW recipe yields a comparable or slightly lower overhang ratio than the 4 kW recipe. Similar results are for the 2 mTorr case. 2 mTorr recipes are overall better than the 5 mTorr ones. The effect of power is probably due to the increased ion flux to the substrate. Whether even higher power can further improve the trench deposition is subjected to some future study. The advantage of lower pressure is from the less gas scattering and thus more directional Cu neutral flux.
8.2.2 Trench Deposition using MPP Solo

A batch of MPP Solo recipes with different pulse-on time \( \tau_{on} \) (8 to 16 µs) is used for the deposition on patterned wafers. The other parameters are 10 µs for pulse-off time, 450 V for the charging voltage, 5 mTorr for the pressure, and average power of 4 kW. The SEMs of the 70 nm and 80 nm trenches after deposition are compared in Figure 8.24. 10-8 marked in the SEM means that the micropulses used are 10 µs off and 8 µs on. It can be seen that 10-8 pulse recipes results in obvious overhang buildup in the 70 nm trenches. The spaces are clearly wider at the top than at about half the height. Deposition using 10-16 pulses has the widest openings. An improvement by using longer \( \tau_{on} \) can also be seen from the 80 nm trenches. There is no obvious overhang in 80 nm trenches by 10-16 pulses. The bad trenches by 10-14 pulses are actually not because of the deposition but due to the inferior trench structures even before the deposition as shown in Figure 8.24 (e). The SEM of such collapsed trenches before deposition is not shown here.
Figure 8.24 SEMs of the 70 and 80 nm trenches after Cu deposition by MPP Solo at 5 mTorr. Different pulse-on time from 8 to 16 µs (a-d) are used. (e) Bad 80 nm trench structures used in the 10-14 recipe.

Other parameters such as the pressure are tried. Figure 8.25 shows SEMs of the trenches deposited at 1, 2, and 5 mTorr. 10-16 pulses and 450 V of $V_{ch}$ are used. 2 mTorr clearly shows an
improvement in both the 70 and 80 nm trenches. The 1 mTorr sample seems to be quite conformal as well but the bright glow near the top opening makes it hard to find the exact widths.

Figure 8.25 SEMs of the 70 and 80 nm trenches after Cu deposition by MPP Solo at 1, 2, and 5 mTorr. Recipes are $\tau_{\text{off}}$ 10 µs, $\tau_{\text{on}}$ 16 µs, $V_{\text{th}}$ 450 V, 1-5 mTorr.

The ratio of overhang to deposition thickness is then determined for each 70 nm trench sample. Figure 8.26 shows the comparison between the samples by DCMS (4 and 16 kW) and those by MPP Solo ($\tau_{\text{on}}$ = 8, 12, 14, 16 µs) at the same pressure of 5 mTorr. Smaller overhangs and thus more conformal depositions are achieved by MPP Solo with longer $\tau_{\text{on}}$. If recalling the Cu ionization fraction measurement (Figure 8.4 and Figure 8.10), an increase of $\tau_{\text{on}}$ from to 16 µs in MPP Solo increases the Cu IF to about 25%. In DCMS, the Cu IFs are about 10% and 19% for 4 and 16 kW. So the improvement of the trench deposition is very likely because of the increased Cu ionization fraction.
Figure 8.26 Comparisons of the overhang/dep ratios in trenches deposited by DCMS and MPP Solo sputtering with different $\tau_{on}$.

Figure 8.27 shows the effect of pressure on the overhang formation during MPP Solo operation. A lower pressure of 2 mTorr does show a possible improvement as compared with 5 mTorr deposition. The overhang/dep ratio at 2 mTorr (9.6%) is comparable to those by DCMS (10.7% and 9.9% for 4 and 16 kW) within the range of error.

Figure 8.27 Overhang/dep ratios in trenches deposited by MPP Solo at different pressure. Recipes are $\tau_{off}$ 10 $\mu$s, $\tau_{on}$ 16 $\mu$s, $V_{ch}$ 450 V, 1-5 mTorr.
8.2.3 Trench Deposition using MPP Cyprium

MPP Cyprium is then applied for the deposition on patterned wafers. The pulse profiles with different $\tau_{on}$ of 8, 10, 12, 14, and 16 $\mu$s and a fixed $\tau_{off}$ of 10 $\mu$s are used. Another pulse profile of 6-10 is also included especially for its higher duty ratio. The charging voltage is 350 V and the pressure is 5 mTorr. The average power varies but the total deposition thickness for each sample is kept the same. 10-16 pulse profile is also used at 2 mTorr.

The SEMs of the 70 nm and 80 nm trenches after these depositions are compared in Figure 8.28. Just by looking at the SEMs, this batch of depositions seems to have an overall conformal deposition. All the 80 nm trenches are wide open with no obvious overhang. For the 70 nm trenches, except for the 10-8 pulse profile, the overhang seems to be well controlled and the step coverage in the middle to bottom range is quite thick and uniform too. Calculations of the overhang/dep ratios further confirm such impression. Figure 8.29 shows the overhang ratios for all the tested recipes and the ratios for the DCMS trenches. At 5 mTorr, overhangs keep decreasing by using longer $\tau_{on}$ and finally 6-10 pulse profile. An improvement as compared with the DCMS is obvious. At 2 mTorr, 10-16 pulse profile also results in more conformal deposition than the DC recipes do.

Such improvements may also be correlated with the Cu ion flux during deposition. As shown in Figure 8.15, the Cu IF increases with longer $\tau_{on}$ and the 6-10 profile has a Cu IF of 20%. However, there seem to be other contributing factors too, especially considering the Cu IF in 16 kW DC is not necessarily much lower. Some further study may be needed to clarify this.
Figure 8.28 SEMs of the 70 and 80 nm trenches after Cu deposition by MPP Cyrium. $V_{ch}$ is 350 V and pressure is 5 mTorr except for 2 mTorr in (f).
8.2.4 Trench Deposition using HiPIMS

Finally, Huettinger HiPIMS plasma generator is employed for the trench deposition. Based on the Cu IF measurements, recipes with high Cu IF are chosen for the deposition, which include 50 µs pulses with $V_{ch}$ of 1400 V and 1600 V, 100 µs pulses with $V_{ch}$ of 1000 V and 1400 V, and 150 pulses with $V_{ch}$ of 1000 V. The pressure of 5 and 2 mTorr are used.

The SEMs of the deposited trenches at 5 mTorr are shown in Figure 8.30. An obvious improvement from the first recipe of 1400 V, 50 µs can be seen by the rest recipes. But still almost all the recipes seem to generate slight overhangs in the 80 nm trenches. By using lower pressure of 2 mTorr, obvious improvement of the conformal deposition can be seen (Figure 8.31). All the 80 nm trenches are widely open.
Figure 8.30 SEMs of the 70 and 80 nm trenches after Cu deposition by HiPIMS at 5 mTorr.
The overhang/dep ratios are then determined from the 70 nm trenches for a better comparison. At 5 mTorr (Figure 8.32 a), the overhang ratios are plotted and compared with the DCMS data. The corresponding Cu IFs are also included in the figure. For HiPIMS, as the IF increases, the overhang ratio decreases. The recipe of 1400 V, 100 µs, 50 Hz slightly improves the trench deposition from the DCMS even though it has so far the highest IF being measured. The other recipes with IFs of about 20% actually do not show improvement. At 2 mTorr (Figure 8.32 a), an increased IF also leads to a reduced overhang ratio for the HiPIMS. Slight improvement from DCMS is achieved by using recipes of 1000 V, 150 µs, 90 Hz and 1400 V, 100 µs, 90 Hz.

The HiPIMS recipes tested so far do not significantly improve the trench deposition as compared with DC and the MPP sputtering, even though the Cu IFs are quite high according to the measurements. The discrepancy may lie in the fact the Cu IFs are measure on the QCM biased at -40V while the depositions are performed on grounded wafers. It has been previously observed that if
the QCM is grounded, the ions collected are reduced. During HiPIMS discharge the presheath extends to the entire chamber at the beginning and it takes time for it to collapse and for the plasma potential to evolve from negative to positive. For DC and MPP which is long enough, the plasma potential is positive so ions have no problem reaching the substrate. If the deposition can be done with a negative bias in HiPIMS, more noticeable improvement may be achieved.

Figure 8.32 HiPIMS overhang to deposition thickness ratio compared with ionization fraction. (a) 5 mTorr, (b) 2 mTorr.
8.3 Conclusions

For the ultimate test of deposition on patterned wafers, the INOVA hollow cathode magnetron is chosen. The idea is by combining two different iPVD techniques together, a higher ionization may be achieved. All three types of power supplies, DC, MPP and HiPIMS are first subject to the plasma characterization, both to study the discharge mechanisms on HCM and to develop potentially good recipes with high Cu ionization fractions. The typical plasma density on substrate level achieved by DC is quite low, e.g. \(10^{17}\) m\(^{-3}\) for up to 16 kW and similar for MPP and HiPIMS typically around \(4 \times 10^{18}\) m\(^{-3}\). The DC Cu ionization fractions in the deposition flux increase with higher power and lower pressure but are typically between 10% and 20%. Higher ionization fractions up to 25% can be achieved using MPP even at low average powers. HiPIMS is able to get a Cu IF as high as 30%. It can be seen that both MPP and HiPIMS are able to enhance the ionization as compared with DC sputtering. However, this fraction is not even as high as in the 200 Gauss configuration in planar magnetron. The very strong magnetic field confinement in HCM may have caused inefficient ion extraction and thus limit the further improvement of ionization fractions.

Patterned wafers with narrow trenches of 70 nm are deposited with in HCM using all three types of sputtering. The deposited trenches are imaged using SEM, and the conformality of the film deposition is examined by using a parameter defined as the ratio of the overhang to the deposition thickness. Smaller overhang is formed at when using a higher power, with the overhang ratio decreases from 14% at 4 kW to 13% at 16 kW at 5 mTorr. This is believed to originate from the higher fraction of Cu ions. The overhang also reduces with a lower pressure, to about 10% at 2 mTorr (16 kW). This is due to more directional neutral flux. The MPP Solo model is seen to improve the trench deposition by using longer micro-pulse on-time. The overhang ratio is reduced to 12% at 5 mTorr. The improvement is associated with a higher Cu ionization fraction as well. More significant improvements have been seen using the MPP Cyprium model. Low overhang ratio of 8% at 5 mTorr is achieved when using some good recipes. HiPIMS, however, only shows slight improvement. It is believed that the HiPIMS plasma potential near the substrate during the pulse may be negative. By biasing the patterned wafer more negatively, a larger amount of ions can arrive to further improve the
trench deposition.

Based on all the tests above, it can be concluded that both MPP and HiPIMS increase the Cu ionization fraction in the deposition flux as compared with the normal DC magnetron sputtering, and the conformality of the trench deposition can be improved. It should be noted that there is still room to further improve the trench deposition here. The other parameters such as the pressure, the extra electromagnetic field, and the substrate bias can all be tuned for this purpose. The scope of the current study is mainly focused on verifying the potential of the pulsed techniques for interconnect metallization, rather than developing recipes that will immediately work on the current node structures. The pulsing techniques, based on the current study, should definitely be further studied, not only at CPMI, but also using a state-of-art PVD system along with extensive process development. Several suggestions can be made for such studies. 1) The magnetic confinement in HiPIMS should be considered as a factor. A stronger confinement does not necessarily yield good performance because of the possible lower ion extraction efficiency. Recall the tests in the 200 to 800 Gauss magnetron configurations, more than 60% of Cu ionization is achieved at 200 Gauss. There should be an optimal magnetic field strength to give the highest fraction of Cu ions. 2) As mentioned above, a negative bias on the substrate may help the ion collections. This is not guaranteed to work thought, since the biased wafer may lower the plasma potential further. 3) A reversed bias on the target may lead to substantial improvement by pushing ions back to the substrate.
Chapter 9  Summary

In this work, high power impulse magnetron sputtering (HiPIMS) and its derivative, modulated pulsed power (MPP) magnetron sputtering, are systematically studied and compared. Their plasma properties and metal ionization fractions are characterized using various pulsing and discharge parameters. Depositions on patterned wafers are performed to evaluate their potential for interconnect metallization application. Time- and spatially-resolved plasma diagnostics are further performed to investigate the physical mechanisms involved in the pulsed plasma generation, evolution, and plasma transport. Specially designed experiments and plasma modeling are used to further understand some key features during HiPIMS, such as the self-sputtering process.

HiPIMS Discharges are fundamentally studied on the planar magnetron. Very high peak current up to 750 A can be achieved. Increasing the charging voltage, pulse duration, and discharge pressure can effectively increase the pulse current, though too long pulse duration may lead to a saturation of the current. The pulse repetition frequency shows little effect. Triple Langmuir probe is employed to study the temporal behaviors of the HiPIMS plasmas. Electron temperature ($T_e$) is observed to decrease dramatically right after the ignition and continue to decrease to throughout the pulse. Electron density ($n_e$) gradually increases during the pulse to about $5 \times 10^{17}$ m$^{-3}$. The initial high $T_e$ is from the high energy electrons escaping the magnetic field confinement. Through the ionization process, electrons lose energies, get better confined and gradually build up a high density plasma in the vicinity of the target. Higher charging voltage and longer pulse duration are seen to dramatically increases $n_e$ while reduces $T_e$.

The time-resolved measurements reveal an obvious plasma expansion with a high density peak moving from the target toward the substrate. It takes about 150 $\mu$s for the expansion peak to reach the substrate. The electron density $n_e$ increases to about 20 times higher as compare with that during the pulse. Therefore the plasma expansion is important for deposition processes. It is believed that a high enough $n_e$ in the confined plasma leads to an enhanced plasma diffusion across the magnetic field. The higher is the $n_e$, the earlier the diffusion starts, and the faster the expansion will be, as observed when varying the charging voltage and pulse duration. A higher pressure leads to a slower expansion because of scattering.
Three-dimensional (3D) triple Langmuir probe measurements allow mapping the plasma density as time evolves to visualize the plasma transport. Based on the 3D measurements, it is found that the plasma transport not only depends on the pulsing and discharge parameters such as the charging voltage, pulse duration and pressure, but is also strongly affected by the magnetic field. With an increased magnetic field strength from 200 Gauss to 800 Gauss, the plasma expansion becomes faster. A large electric potential drop is observed in the presheath and extends into the bulk plasma region. This potential drop remains large for a long time in the 800 Gauss configuration. The corresponding electric field promotes the electron drifting. At the same time, the higher density in the 800 Gauss configuration promotes the diffusion process across the B field. The electric potential distribution leads to the plasma expansion changing from close to isotropic to more directional towards the substrate.

Cu ionization fractions (IF) are measured on the substrate level using HiPIMS. Up to 60% has been achieved, much higher than the DC magnetron sputtering. It basically increases with higher charging voltage and longer pulse length due to higher plasma densities. A stronger B field, in spite of a higher plasma density, produces lower Cu ionization. This is attributed to a lower ion extraction efficiency due to the plasma potential distribution.

Plasma fluxes to the cathode are for the first time directly measured through an orifice in the race track region. The ion fluxes are measured on a negatively biased grid. It is seen that the ion current increases abruptly after a certain time delay instead of increasing smoothly from the pulse beginning. This time delay depends on recipes, for example, being longer for lower charging voltages. This suggests a mechanism that plasma is ignited only in a long strip where the B field is strong and drifts toward the weak-B region. A higher charging voltage allows plasma to ignite in a larger region and drift faster. Fluxes of Cu atoms and Cu\textsuperscript+ ions are measured using Si witness plates and then compared with the total ion flux. A large ratio of Cu\textsuperscript+ to Ar\textsuperscript+ is determined as a direct evidence for the enhanced self-sputtering during HiPIMS.

To provide more insights into the development of Cu ion and Ar ion species, a time-dependent model is built to describe the ionization region where plasma is confined by magnetic field. The essential processes such as plasma-target interactions, electron collision ionizations, and gas rarefaction are incorporated in the model. Previously-measured discharge voltage and current along
with several other parameters based on the experimental data are used as input. The test results of the model show the capability to predict the temporal development of the electron density, the degrees of ionization for Cu and Ar, and the ratio of Cu ions to Ar ions. Based on the model results, strong gas rarefaction is shown to take place while the self-sputtering by Cu takes over. The sensitivity of the model on various input parameters such as the geometry of the ionization region and the electron temperature are investigated.

Magnetic field configurations are modified specifically for the high power impulse magnetron sputtering (HIPIMS). The race track pattern is varied to optimize the target utilization and the downstream plasma uniformity. Attempts are made to spread the erosion evenly on the target, but the discharge currents are relatively low due to the non-closed race tracks. A closed path for electrons to drift along is still essential in HIPIMS. The configuration of wider race track generates a higher pulse current, and extends the intense plasma coverage on the substrate. A spiral-shaped magnetic field configuration is able to generate high pulse current, achieve a downstream plasma with superior uniformity, and yield a better target utilization even without the assistance of magnet rotation.

MPP discharge is studied on the planar magnetron, using both Solo and Cyprium models. By adjusting the arrangement of micro-pulses, custom pulse shapes are achieved. The I-V characteristics show multiple stages including one with high discharge currents. The Cyprium model exhibits a distinctive feature that the voltage and current strongly oscillate. Similar plasma densities during the pulses are achieved in MPP as in HiPIMS, though there is no obvious high-density expansion peak. Stronger plasmas can be achieved using longer micro-pulse on-time, shorter off-time and higher charging voltage. The deposition rates are about twice higher than those in HiPIMS at the same average power. The ionization fractions are shown to increase with longer micro-pulse on-time, higher charging voltage, etc. It also has a similar dependence on the B field strength. Overall the ionization fractions by MPP are much lower as compared with HiPIMS. This is likely because of the too short off-time (on the order of 10µs) prevents the ion release, even for the Cyprium model with designs of reducing voltage between micro-pulses.

All three types of power supplies, DC, MPP and HiPIMS are tested on a 200 mm hollow cathode magnetron before being applied for the patterned wafer tests. DC discharge is shown to have a low plasma density of around $10^{17}$ m$^{-3}$ on the substrate even at very high input power. The Cu
ionization fractions in the deposition flux increase with higher power and lower pressure but are typically between 10% and 20%. MPP plasma generators greatly increase the $n_e$ during pulse to about $4 \times 10^{18}$ m$^{-3}$. High ionization fractions up to 25% can be achieved even at low powers. HiPIMS is able to get a comparable peak density as MPP with the similar plasma expansion behaviors as seen in the planar magnetron tests. The Cu ionization fractions are measured to be as high as 30%. The very strong magnetic field confinement in HCM may have caused inefficient ion extraction and thus limit the further improvement of ionization fractions.

Patterned wafers with narrow trenches of 70 nm are deposited with in HCM using all three types of sputtering. The deposited trenches are imaged using SEM, and the conformality of the film deposition is examined by using a parameter defined as the ratio of the overhang to the deposition thickness. Smaller overhang is formed at when using a higher power, with the overhang ratio decreases from 14% at 4 kW to 13% at 16 kW at 5 mTorr. This is believed to originate from the higher fraction of Cu ions. The overhang also reduces with a lower pressure, to about 10% at 2 mTorr (16 kW). This is due to more directional neutral flux. The MPP Solo model is seen to improve the trench deposition by using longer micro-pulse on-time. The overhang ratio is reduced to 12% at 5 mTorr. The improvement is associated with a higher Cu ionization fraction as well. More significant improvements have been seen using the MPP Cyprium model. Low overhang ratio of 8% at 5 mTorr is achieved when using some good recipes. HiPIMS, however, only shows slight improvement. It is believed that the HiPIMS plasma potential near the substrate during the pulse may be negative. By biasing the patterned wafer more negatively, a larger amount of ions can arrive to further improve the trench deposition. It should be noted that these tests are not for developing processes that will yield instant success on the small 1X structures in the current node. Instead, they are designed to show the potential of the pulsed techniques for interconnect metallization. And the above tests do present some obvious positive signals as compared with the normal DC magnetron sputtering.

In all, the current study investigates the HiPIMS/MPP as a new alternative technique for barrier/seed layer deposition, which is urgently needed for the next generation. The HiPIMS and MPP may prove successful as the future iPVD technique for this application. Overall advantages of HiPIMS and MPP over the normal DC sputtering are demonstrated through both plasma studies and
patterned wafers tests. A substantial increase of the metal ionization fraction is believed to be the key. The promoted self-sputtering results in a lower Ar$^+$ fraction, which is also desirable for the seed deposition. This study also concentrates on exploring some puzzling HiPIMS discharge mechanisms. A great value of this work is to provide a direct evidence of the self-sputtering effect by measuring the fluxes through a hole in the target. High fraction of Cu$^+$ flux is determined as well as predicted using an ionization region model. The low ion extraction efficiency is a general problem which leads to the deposition rate loss in HiPIMS. The low ion extraction efficiency is found to be caused by the non-flat plasma potential distribution. One possible solution is to properly reduce the magnetic field confinement. The orientation of the plasma expansion can be changed, e.g. from isotropic to directional, which open up for new possibilities of an independent control to improve the uniformity. All these new findings lead to a further understanding of the discharge and better control of the HiPIMS. It is also suggested the magnetic field configuration to be built specifically for HiPIMS. A thorough comparison between HiPIMS and MPP is performed, providing some useful guideline information should a choice need to be made for various applications.
Chapter 10  Future Work

While the current work provides a systematic comparison of two types of advanced high power pulsed techniques and proves their potential for improving interconnect metallization, there is still more work to be done. Several points are listed below. These can either help further understand the HiPIMS/MPP discharge mechanisms or facilitate the applications of the pulsed techniques, or even improve the plasma generators themselves.

The HiPIMS plasma study shows that electric potential distribution plays a critical role in both the plasma transport and the ion extraction. A direct measurement of the plasma potential instead of the floating potential will provide more accurate information of the electric fields in the space. The plasma potential can be measured using an emissive probe. With proper design, such measurements can be made time-resolved and spatially-resolved. The information on the time evolution and the spatial distribution of the plasma potential will help further explain some of the observations. The underlying physics of the plasma potential distribution itself should be investigated, along with a study on the contributing factors. In the end, it is desirable to be able to control the plasma potential distribution to optimize the processes.

Further optimization of the trench deposition should be continued. Improving the ion extraction efficiency seems to be crucial. On one hand, we know it is affected by the plasma potential distribution. So the influences of different pulsing and discharge parameters and the magnetic field should be studied to control the plasma distribution during the pulse. On the other hand, it is worth trying to control the “after-pulse” plasma by reversing the bias on the target after each pulse. As found in the plasma transport experiments, there is a high-density plasma in the magnetic confinement region even at the end of the pulse. Getting all the ions out to the substrate may give a substantial increase of ion flux. Without the reversed bias, however, the negative sheath will still attract the ions to the target. This has been verified by the potential measurement in 800 Gauss configuration. The potential drop remains for another 100 \( \mu \)s after pulse ends. This may not help MPP discharge since the macro-pulse is much longer to see much effect on average. However, for MPP itself, the idea of reducing the voltage to zero can be further improved. The advantage of MPP Cyprium model over Solo model, though not completely sure, may have some interrelationship with this operation.
mechanism. Other than the modifications of the power supplies, it is worth studying to bias the substrate to test if HiPIMS can further improve the trench deposition. After all, it has yielded the highest Cu IF with -40 V on the QCM.

A continued study using the through-target diagnostics should be performed. The test assembly was designed to work with QCM for distinguishing the metal atom and ion species. A differential pumping can be included to reduce the gas scattering. This will yield more straightforward and more accurate results on the Cu\(^+\)/Ar\(^+\) ratio, etc. Using two grids, the ion energy distribution can be measured. This will provide important information on the sheath/presheath structures, considering that ions generated at different plasma potential will have different energy reaching the target. The orifice can be moved to different regions in the race track with different B field strength, to confirm the conjecture of moving plasma strip.

More studies of HiPIMS or MPP discharge should be done on small-size magnetrons for the study of underlying physics. While the tests on large industrial-size chambers are closer to the real application requirements, high power densities are hard to achieve on large magnetrons due to the limitation by the rated power or current of our generators. Many critical features of HiPIMS, unfortunately, only happen at very high power densities, such as the localization of ionization zones and high degree of self-sputtering.

A continual effort on the HiPIMS modeling is needed. Because of the unique and unknown mechanisms involved in HiPIMS or MPP, the need for detailed modeling is greater than ever. The current model is based on many assumptions, which is good for predicting some core processes. But in order to understand the more complex mechanisms, such as the plasma potential distribution and evolution, the localization of ionization, many assumptions have to be removed with new effects added. The ionization zone won’t be zero-dimension, and will evolve over time and change with the magnetic field topology. The electron energy distribution will have to be derived from the power balance. The loss of electrons by cross-B diffusion will need to be included. More species such as excited atoms, hot Ar atoms should be included as well.
References


J. Sporre, Diagnosis of the flux emanating from the intermediate focus of an extreme ultraviolet light lithography source, PhD thesis, University of Illinois at Urbana-Champaign, 2013.