Chapter 4

Discharges, Plasmas, and Ion-Surface Interactions
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Physical Vapor Deposition (PVD)

- Evaporation caused by absorption of thermal energy is not the only way to induce atoms to leave a liquid or solid surface. (Evaporation)
- Atoms can also be ejected or sputtered from solids at room temperature by bombarding their surfaces with energetic ions. (Sputtering)
- In either case the emitted atoms traverse a reduced pressure ambient and deposit atomistically on a substrate to form a film. Because physical means are primarily involved in producing films, both are known as physical vapor-deposition (PVD) processes.
Schematics of a DC Sputtering System

Several kilovolts

Sputtering System for Metal Deposition

- The system consists of a chamber, inside which is a pair of **parallel metal plates** as electrodes.
- One of parallel metal is the **cathode** or **target** of the metal to be deposited.
  - It is connected to the **negative terminal** of a DC power supply and typically, several kilovolts are applied to it.
- Facing the cathode is the **substrate** or **anode**, which may be grounded, biased positively or negatively, heated, cooled, or some combination of these.
- After **evacuation of the chamber**, a working gas, typically **argon**, is introduced and serves as the medium in which an electrical discharge is initiated and sustained.
  - Gas pressures usually range from a few to a hundred millitorr.
- After a visible **glow discharge** is maintained between the electrodes, it is observed that a current flows, and metal from the cathode deposits on the substrate.
Schematics of a Sputtering

Target/Cathode is not necessarily at the top.

Sputter | Prof. Igor Lubomirsky's Lab
Weizmann Institute of Science
https://www.weizmann.ac.il/materials/igorl/sputter
Microscopic Description of Sputtering

- Positive gas ions in the discharge strike the cathode and physically eject or sputter target atoms through momentum transfer to them.
- These atoms enter and pass through the discharge region to eventually deposit on the growing film.
- In addition, other particles (secondary electrons, desorbed gases, and negative ions) as well as radiation (X-rays and photons) are emitted from the target.
- The electric field accelerates electrons and negatively charged ions toward the anode substrate where they impinge on the growing film.
Sputtering v.s. Evaporation

Sputtering
• An ionized gas or plasma
• Active electrodes that participate in the deposition
• Low temperature processing
• Eject of atoms by momentum transfer
• The glow-discharge plasma is very busy and not easily modeled

Evaporation
• Rarefied-gas environment (higher vacuum)
• Passive electrode
• High temperature processing
• Thermally induced evaporation
• Predictable rarefied-gas behavior (molecular flow)
DC and RF Sputtering Systems

Several kilovolts are applied and gas pressures usually range from a few to a hundreds millitorr.
Application of Plasma

• In the past few decades, advances in our understanding of the physics and chemistry of ionized gases has led to the widespread adoption of plasma technology for the deposition and removal (etching) of thin films as well as the modification of surfaces in a diverse variety of technologies.
• Microelectronics applications have been the main technological driver in this regard.
• In addition, there are critical plasma processing operations in the automotive, optical coating, biomedical, information recording, waste management, and aerospace industries.
Scientific Issues in Glow Discharge Systems

Regardless of the plasma process, however, roughly similar discharges, electrode configurations, and gas/solid interactions are involved.

1. Initiating and sustaining discharges
2. The dynamical behavior of the charged and neutral species
3. Species interaction within the plasma
4. Both inert gas discharges and the more complex chemically reactive plasmas
5. Ion interactions with the cathode and film or substrate surfaces
6. Fundamental physics of sputtering
7. Ion-induced modification of growing films
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Plasma

- A *quasi-neutral gas* that exhibits a *collective* behavior in the presence of applied electromagnetic fields.
- Weakly ionized gases consisting of a collection of electrons, ions, and neutral atomic and molecular species.
- This definition is broad enough to encompass the spectrum of space and man-made plasmas extending from stars to fusion reactors.

Types of plasmas (by density of charges)

- Stars (density $n < 10^7 \text{ cm}^{-3}$)
- Solar winds (density $n < 10^7 \text{ cm}^{-3}$)
- Coronas (density $n < 10^7 \text{ cm}^{-3}$)
- Ionosphere (density $n < 10^7 \text{ cm}^{-3}$)
- Glow discharge (density $n = 10^8 \sim 10^{14} \text{ cm}^{-3}$)
- Arcs (density $n = 10^8 \sim 10^{14} \text{ cm}^{-3}$)
- High-pressure arc (density $n \sim 10^{20} \text{ cm}^{-3}$)
- Shock tubes (density $n \sim 10^{20} \text{ cm}^{-3}$)
- Fusion reactors (density $n \sim 10^{20} \text{ cm}^{-3}$)
Plasmas consist of freely moving charged particles, i.e., electrons and ions. Formed at high temperatures when electrons are stripped from neutral atoms, plasmas are common in nature. For instance, stars are predominantly plasma. Plasmas are a “Fourth State of Matter” because of their unique physical properties, distinct from solids, liquids and gases. Plasma densities and temperatures vary widely.
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Discharge

1. We have seen that application of a sufficiently high DC voltage between metal electrodes immersed in a low-pressure gas initiates a discharge (放電).

2. In essence, the discharge reflects a gaseous breakdown that may be viewed as the analog of dielectric breakdown in insulating solids.

3. Their dielectrics conduct electricity at critical applied voltages. In gases, the process begins when a stray electron near the cathode carrying an initial current $i_o$ is accelerated toward the anode by the applied electric field ($\mathcal{E}$).
Principal Glow Discharge Mechanism by Biased Parallelplate

1. A stray electron near the cathode carrying an initial current \( i_0 \) is accelerated toward the anode by the applied electric field (\( \varepsilon \)).

2. After gaining sufficient energy the electron collides with a neutral gas atom (A), e.g., Ar, converting it into a positively charged ion (A\(^+\)), i.e., \( e^- + A \rightarrow 2e^- + A^+ \).

3. Two electrons are generated and are accelerated and bombard two additional neutral gas atoms, generating more ions and electrons, and so on.

4. Meanwhile, the electric field drives ions in the opposite direction.

5. Ions collide with the cathode, ejecting, among other particles, secondary electrons.

6. Secondary electrons also undergo charge multiplication. (step 2)

7. The effect snowballs until a sufficiently large avalanche current ultimately causes the gas to breakdown.

![Diagram of glow discharge mechanism](http://webhost.ua.ac.be/plasma/pages/glow-discharge.html)
Avalanche Breakdown

\[ e^- + A \rightarrow 2e^- + A^+ \]

1. In order for breakdown to occur, the distance (d) between electrodes must be large enough to allow electrons to incrementally gain the requisite energy for an ionization cascade.

2. Also, the electrodes must be wide enough to prevent the loss of electrons or ions through sideways diffusion out of the inter-electrode space.

---

**Anode (+)**

---

**Cathode (−)**
Townsend Equation

\[ i = i_o \frac{e^{\alpha d}}{1 - \gamma_e (e^{\alpha d} - 1)} \]

- **i**: charge current
- **\( \alpha \)**: Townsend ionization coefficient
- **\( \gamma_e \)**: Townsend secondary-electron emission coefficient
- **d**: Distance between electrodes

Townsend ionization coefficient (\( \alpha \)) represents the probability per unit length of ionization occurring during an electron-gas atom collision. Townsend secondary-electron emission coefficient (\( \gamma_e \)) is the number of secondary electrons emitted at the cathode per incident ion.
Townsend Ionization Coefficient

\[ \alpha = \frac{1}{\lambda} e^{\frac{-E_i}{q \varepsilon \lambda}} \]

- \( \alpha \): Townsend ionization coefficient
- \( q \): Electron charge
- \( \lambda \): traveling distance
- \( E_i \): Ionization potential
- \( \varepsilon \): Applied electric field

For an electron of charge \( q \) traveling a distance \( \lambda \), the probability of reaching the ionization energy \( E_i \) is

\[ \alpha = \frac{1}{\lambda} e^{\frac{-E_i}{q \varepsilon \lambda}}. \]

We may associate \( \lambda \) with the intercollision distance or mean free path in a gas. Since \( \lambda \sim P^{-1} \), we expect \( \alpha \) to be a function of the system pressure.
Mean-Free Path

\[ A = \pi d^2 \]

\[ n = \frac{\#}{V} = \frac{N_A}{RT} \frac{RT}{P} \]

\[ \frac{L}{A \cdot L \cdot n} = \lambda \]

\[ \lambda = \frac{RT}{\pi N_A d^2 P} \propto P^{-1} \]
Derivation of Townsend Equation

(a) The total current $I = I_-(x) + I_+(x)$, a sum of electron and ion components, is constant, i.e., independent of $x$ and time.

(b) At the cathode $I_-(0) = I_o + \gamma e I_+(0)$, with $I_o$ a constant.

(c) Across the gap, $d[I_-(x)]/dx = \alpha I_-(x)$, i.e., $I_-(x) = I_-(0) \exp \alpha x$.

(d) At the anode, $I_+(d) = 0$. 
Breakdown Voltage

Breakdown is assumed to occur when the denominator of the Townsend equation is equal to zero, i.e., $\gamma_e (e^{\alpha d} - 1) = 1$, for then the current is infinite.

Townsend equation \[ i = i_o \frac{e^{\alpha d}}{[1 - \gamma_e (e^{\alpha d} - 1)]]} \]

\( i \): charge current  
\( \alpha \): Townsend ionization coefficient  
\( \gamma_e \): Townsend secondary–electron emission coefficient  
\( d \): Distance between electrodes  
\( V_i \): Ionization potential

\[
\lambda = \frac{RT}{\pi N_A \phi^2 P} \propto P^{-1} \\
\alpha = \frac{1}{\lambda} e^{-\frac{E_i}{q \varepsilon \lambda}} = \frac{1}{\lambda} e^{-\frac{V_i}{\varepsilon \lambda}} \\
\phi: Diameter of a gas molecule
\]

Paschen’s Law

\[ V_B = \frac{APd}{\ln(Pr) + B} \]

\( A, B \): constants
• At low values of $Pd$ there are few electron-ion collisions and the secondary electron yield is too low to sustain ionization in the discharge.
• At high pressures there are frequent collisions, and since electrons do not acquire sufficient energy to ionize gas atoms, the discharge is quenched.
• In between, at typically a few hundred to a thousand volts, the discharge is self-sustaining.
• Practically, however, in most sputtering discharges the $Pd$ product is well to the left of the minimum value.

$$V_B = \frac{APd}{\ln(Pd) + B}$$
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Types and Structures of Discharges

Townsend discharge
A tiny current flows initially due to the small number of charge carriers in the system. With charge multiplication, the current increases rapidly, but the voltage, limited by the output impedance of the power supply, remains constant.

Normal glow
When enough electrons produce sufficient ions to generate the same number of initial electrons, the discharge becomes self-sustaining. The gas begins to glow now and the voltage drops accompanied by a sharp rise in current. Initially, ion bombardment of the cathode is not uniform but concentrated near the cathode edges or at other surface irregularities. As more power is applied, the bombardment increasingly spreads over the entire surface until a nearly uniform current density is achieved.

Abnormal discharge
A further increase in power results in both higher voltage and cathode current-density levels. The abnormal discharge regime has now been entered and this is the operative domain for sputtering and other discharge processes such as plasma etching.

Arc
At still higher currents, the cathode gets hotter. Now the thermionic emission of electrons exceeds that of secondary-electron emission and low-voltage arcs propagate. Arc is a self-sustained discharge that supports high currents by providing its own mechanism for electron emission from negative or positive electrodes.
Current–voltage \((i - V)\) characteristics of direct current (dc) electrical discharge

\(V_b\) is the breakdown voltage,

\(V_n\) is the normal operating voltage, and

\(V_d\) is the operating voltage of arc discharge.

http://www.spectroscopynow.com/Spy/pdfs/jwfeed/sample_0471606995.pdf
Glow discharge in a low-pressure tube caused by electric current. Performed by Prof. Oliver Zajkov at the Physics Institute at the Ss. Cyril and Methodius University of Skopje, Macedonia.
http://www.exo.net/~pauld/origins/glowdisharge.html

Sprite light (精靈放電) in the atmosphere (left) and in a laboratory glow discharge tube (right). In both cases, the light near the positive (anode) end is red and arises from the collisional excitation of neutral nitrogen molecules by free electrons. Also in both cases, the light near the negative (cathode) end is blue and arises from the collisional excitation of $\text{N}_2^+$ ions by free electrons.
Regions in the DC Glow Discharge Tube

A glass tube, about *16 inches long* and *1 1/2 inches in diameter*, is hermetically sealed at both ends. Two metal probes are fused into the tube at each end. The physicist applies a potential of a few thousand volts across both probes. With the aid of a vacuum pump he sucks the *air* out of the tube, thus lowering the pressure inside the glass tube.
Regions in the DC Glow Discharge Tube

the air rarefied

http://www.newjerusalemnetwork.net/emmanuel/great_wall3.html
Regions in the DC Glow Discharge Tube

A First of all, a narrow beam of **red light** moves across the inside of the tube, from one electrode to the other. As the pressure continues to fall, the beam of light becomes thicker and soon fills the whole space between the electrodes. Near the one electrode there appears a **blue glow**, and at the other a dark space.

If the physicist continues to rarefy **the air** in the tube, the blue glow turns into a thin **red sheath** of light close to the electrode [identified in the illustration to the top-left as A].

B After a region of darkness, known as 'Crookes' dark space' after its discover, there appears with a well defined edge, the palest part of the electrical discharge, a **faint blue column** of light known as the **negative glow**.

C This blue glow fades gradually into another dark space, which is followed by a weak **red-violent glow** known as the **positive column** or 'plasma.'

D In the final stage of the experiment, **the air** in the tube is rarefied until all the discharge phenomena disappear. Then, however, the right-hand end of the glass glows with a **Green Fluorescent Light**.
Regions in the DC Glow Discharge Tube

Although the general structure of the discharge has been known for a long time, the microscopic details of the charge distributions, behavior, and interactions within these regions are not totally understood.

Cathode

Aston dark
The Aston dark space is very thin and contains both low energy electrons and high energy positive ions, each moving in opposite directions.

Cathode glow
Beyond the Aston dark the cathode glow appears as a highly luminous layer that envelops and clings to the cathode.

De-excitation of positive ions through neutralization is the probable mechanism of light emission here.

Cathode dark (Crookes)
Next to cathode glow is the cathode dark space, where some electrons are energized to the point where they begin to impact-ionize neutrals; other lower energy electrons impact neutrals without ion production.

Because there is relatively little ionization this region is dark.

Most of the discharge voltage is dropped across the cathode dark space.

Cathode dark space is commonly referred to as the cathode sheath.

The resulting electric field serves to accelerate ions toward their eventual collision with the cathode.
Regions in the DC Glow Discharge Tube

Negative glow
Here the visible emission is apparently due to interactions between assorted secondary electrons and neutrals with attendant excitation and de-excitation. During sputtering the substrate is typically placed inside the negative glow before the Faraday dark space so that the latter as well as the positive column do not normally appear.

Faraday dark

Positive column
With fewer ions, the electric field increases, resulting in electrons with energy of about 2 eV, which is enough to excite atoms and produce light. With longer glow discharge tubes, the longer space is occupied by a longer positive column, while the cathode layer remains the same.

Bands of alternating light and dark in the positive column are called striations. Striations occur because only discrete amounts of energy can be absorbed or released by atoms, when electrons move from one quantum level to another. The effect was explained by Franck and Hertz in 1914.

With neon, the Franck–Hertz voltage interval is 18.7 volts, and an orange glow appears near the grid when 18.7 volts is applied. This glow will move closer to the cathode with increasing accelerating potential, and indicates the locations where electrons have acquired the 18.7 eV required to excite a neon atom.

Anode dark
Commonly referred to as the anode sheath.

Anode
Neon atoms have 10 electrons and a ground state of \(1s^2\ 2s^2\ 2p^6\) (see Figure). Due to electron spin-related selection rules, collisions with electrons excite neon atoms from the ground state to the \(2p^5\ 3p\) and \(2p^5\ 4p\) states. When falling back toward the ground state by emitting photons the \(2p^5\ 3s\) state is also allowed. Recall that you can calculate the wavelength of the photons emitted from \(E = \frac{hc}{\lambda}\).
Regions in the Glow Discharge Tube

Cathode dark (Crookes) (Cathode sheath)

Aston dark

Faraday dark

Cathode glow

Negative glow

Positive column

Anode dark (Anode sheath)

Cathode

Anode

Cathode glow

Negative glow

Positive column

Anode glow

Cathode (-)

Anode (+)
Potentials along the Tube

Light intensity

Potential V

Field E

Current

Charge density

Charge density (total)
Regions in the Glow Discharge Tube II

Cathode layer  Negative glow  Positive column

Anode glow

Dark spaces

Crookes  Faraday

Aston  Anode

50 cm at 1 Torr

Figure 4-3 Structure of a DC glow discharge with corresponding potential, electric field, charge, and current distributions.
\[
\frac{dV}{dx} = -E
\]

\[
\frac{dE}{dx} = \frac{1}{\varepsilon} q(N^+ - N^-)
\]

\[N^+ - N^-\]

Figure 4-3 Structure of a DC glow discharge with corresponding potential, electric field, charge, and current distributions.

One can see that where there is not a glowing discharge, we either have a greater potential drop or a very low pressure.
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Plasma Species

1. Plasma is composed of respective densities of electrons ($n_e$), ions ($n_i$), and neutral gases ($n_o$).

2. Electrons and ions have more or less independent velocity distributions with electrons possessing far higher velocities than ions.

3. The plasma is electrically neutral when averaged over all the particles contained within so that $n_e = n_i = n$.

Motion of Species

1. Collisions between neutral gas species essentially cause them to execute random Brownian motion.

2. However, the applied electric field disrupts this haphazard motion because of ionization.

3. If the density of charged particles is high enough compared with the dimensions of the plasma, significant coulombic interaction exists among particles.

4. This interaction enables the charged species to flow in a fluid-like fashion that determines many of the plasma properties.
Degree of gas ionization

1. The degree of ionization \( f_i \) is defined by \( f_i = n_e/(n_e+n_o) \).
2. The degree of ionization \( f_i \) typically has a magnitude of \(-10^{-4}\) in the glow discharges used in thin-film processing.
3. Therefore, at pressures of \(-10^{-4}\) millitorr, a gas density of \( n_o = ~10^{14} \) cm\(^{-3}\), based on the ideal gas law.
4. Hence the electron and ion densities will be about \(10^{10} \) cm\(^{-3}\) each at 25\(^\circ\)C.
5. In high density plasmas, \( f_i \) can reach \(-10^{-2}\) and charge densities more than \(10^{12} \) cm\(^{-3}\).

Reminder:
Plasma species: electron \((n_e)\), ions \((n_i)\), and neutral gas \((n_o)\)
Electron has far higher velocities than ions. \(v_e >> v_i\)
Electrically neutral: \(n_e = n_i\)
Degree of gas ionization: \( f_i = n_e/(n_e+n_o) \)
\( f_i = 10^{-4} \) for glow discharge, and \(10^{-2}\) in high density plasmas.
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Particle Energies and Temperatures

For glow discharges:

• Electron energy $E_e = 1$ to $10$ eV (range of span, typically $2$ eV)
• Effective characteristic temperature $T_e = E_e / k_B = 23000$ K ($k_B = 8.617 \times 10^{-5}$ eV/K)
• Neutral gas energy $E_o = 0.025$ eV, ($T_o = 293$ K)
• Ion energy $E_i = 0.04$ eV ($T_i = 500$ K), acquired from electric field.

Temperature partitioned in different molecular modes:

• In addition, there may be excited species at temperature $T_{ex}$ with energy $E_{ex}$.
• Neutral molecules may become excited by virtue of acquired energy that is partitioned into translational as well as internal vibrational and rotational modes of motion.
• For each of these modes there is a corresponding characteristic temperature.
• E.g., for nitrogen gas plasma at several torr,
  - $T_e \sim 12000K$,
  - $T_o$ (due to molecular translation) $\sim 12000K$,
  - $T_v$ (for molecular vibrational) $\sim 3800K$,
  - $T_{ro}$ (for molecular rotational) $\sim 280K$, respectively.

Low pressure glow discharge is a nonequilibrium or cold plasma.
(if otherwise in equilibrium: $T_i = T_o = T_e = T_{ex} = T_r$ (radiation) = $T_w$ (chamber wall) = $T$)
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Motion in Plasma Species

Charged particle impingement results in an effective electrical current density \( (j) \) given by the product of the particle flux \((\frac{1}{4} \cdot n\bar{v})\) and the charge \((q)\) transported, where the factor of \( \frac{1}{4} \) reflects that fraction of the random motion that is directed at the planar surface.

\[
j = \frac{n\bar{v}}{4} q
\]

\[ \Phi = n\sqrt{\frac{M}{2\pi RT}} \int_0^\infty v_x \exp\left(-\frac{Mv_x^2}{2RT}\right)dv_x = n\sqrt{\frac{RT}{2\pi M}} = \frac{n\bar{v}}{4} \]

\[
\therefore \bar{v} = \sqrt{\frac{8k_B T}{\pi m}}
\]
Velocity of Particles in Plasma

\[ \overline{v} = \sqrt{\frac{8k_B T}{\pi m}} \]

\[ k_B = 1.38 \times 10^{-23} \text{ m}^2 \cdot \text{kg} \cdot \text{s}^{-2} \cdot \text{K}^{-1} \]

\[
\left\{ \begin{array}{l}
\mit{e} = 9.1 \times 10^{-28} \text{ g} \\
T_e (\text{assume}) = 23000 \text{ K}
\end{array} \right.
\] \quad \rightarrow \quad v_e = 9.5 \times 10^7 \text{ cm/s}

\[
\left\{ \begin{array}{l}
m_{i(Ar)} = \frac{40}{6.02 \times 10^{23}} = 6.64 \times 10^{-23} \text{ g} \\
T_i = 500 \text{ K}
\end{array} \right.
\] \quad \rightarrow \quad v_i = 5.2 \times 10^4 \text{ cm/s}
Currents in Plasma

For typical Ar ions, \( v_i = 5.2 \times 10^4 \text{ cm/s} \)
For electrons, \( v_e = 9.5 \times 10^7 \text{ cm/s} \)

If \( n_e = n_i = 10^{10} / \text{cm}^3 \)

\[ j_e \sim 38 \text{ mA/cm}^2 \]
\[ j_i \sim 21 \mu\text{A/cm}^2 \]

The implication of this calculation is that an isolated surface within the plasma charges negatively initially because of the greater electron bombardment. Subsequently, additional electrons are repelled while positive ions are attracted. Therefore, the surface continues to charge negatively at a decreasing rate until the electron flux equals the ion flux and there is no net current.

Surface are charged negatively due to greater electron bombardment. \( v_e > v_i \)
Mobility in an Electric Field

Mobility: velocity per unit electric field \( \mu = \frac{\nu}{\mathcal{E}} \)

Electric force

\[ m \frac{d\nu}{dt} = q|\mathcal{E} + m\left[ \frac{\partial \nu}{\partial t} \right]_{coll} \]

Frictional drag

\[ = q|\mathcal{E} + m\nu \nu \]

Collision frequency

\[ \therefore \left[ \frac{\partial \nu}{\partial t} \right]_{coll} = \nu \nu \]

In the steady state,

\[ \frac{d\nu}{dt} = 0 \rightarrow \nu = \frac{|q|\mathcal{E}}{m\nu} \]

\[ \mu = \frac{|q|}{m\nu} \quad (\therefore \mu = \frac{\nu}{\mathcal{E}}) \]

Typical mobilities for gaseous ions at 1 torr and 273 K range from \( \sim 4 \times 10^2 \text{ cm}^2/\text{V-s} \) (for Xe\(^+\)) to \( 1.1 \times 10^4 \text{ cm}^2/\text{V-s} \) (for H\(^+\)).
Diffusion in an Concentration Gradient

A second kinetic effect involving species motion in plasmas is diffusion, a phenomenon governed by Fick’s Law.

\[
\text{Flux } J = -D \frac{dn}{dx} \quad \text{#/(cm}^2 \cdot \text{s)}
\]

\[
J_e (\text{diffusion}) = -D_e \frac{dn_e}{dx}
\]

\[
J_i (\text{diffusion}) = n_i \mu_i \mathbf{E} - D_i \frac{dn_i}{dx}
\]
Diffusion and Drift

When migrating species move under the simultaneous influence of two driving forces, i.e., diffusion in a concentration gradient \((dn/dx)\) and drift in the applied electric field, we may write for the respective electron and ion particle \textit{fluxes},

\[
J_e = -n_e \mu_e \mathbb{E} - D_e \frac{dn_e}{dx}
\]

\[
J_i = n_i \mu_i \mathbb{E} - D_i \frac{dn_i}{dx}
\]

Charge Neutrality, \( J_e = J_i = J \), \( n_e = n_i = n \)

By equating \( J_e \) and \( J_i \),

\[
\mathbb{E} = \frac{(D_i - D_e)}{n(\mu_e + \mu_i)} \frac{dn}{dx}
\]

An electric field develops because the difference in electron and ion diffusivities produces a separation of charge.
Ambipolar Diffusion Coefficient

\[ \mathcal{E} = \frac{(D_i - D_e)}{n(\mu_e + \mu_i)} \frac{dn}{dx} \]

- An electric field develops because the difference in electron and ion diffusivities produces a separation of charge.
- Physically, more electrons than ions tend to leave the plasma, establishing an electric field that hinders further electron loss but at the same time enhances ion motion.

Substitute this field into either of the flux equations to get an expression for the flux in terms of the concentration gradient,

\[ J_i = n_i \mu_i \frac{(D_i - D_e)}{n(\mu_e + \mu_i)} \frac{dn_e}{dx} - D_i \frac{dn_i}{dx} = - \frac{(D_i \mu_e + D_e \mu_i)}{(\mu_e + \mu_i)} \frac{dn}{dx} \]

- Because of the coupled electron and ion motions we can assign an effective ambipolar diffusion coefficient \( D_a \) to describe the effect.

Ambipolar diffusion coefficient \( D_a = \frac{(D_i \mu_e + D_e \mu_i)}{(\mu_e + \mu_i)} \)

- The magnitude of \( D_a \) lies somewhere between those of \( D_i \) and \( D_e \).
- Both ions and electrons in plasma diffuse faster than intrinsic ions do.
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Electron Motion in Combined Electric Field

- **Parallel Fields**: A magnetic field of strength $B$ is superimposed parallel to the electric field $\mathbf{E}$ between the target and substrate.

  \[ \mathbf{B} \parallel \mathbf{E} \]

- **Perpendicular Fields**: Electric field $\mathbf{E}$ is still normal to the cathode while magnetic field of strength $B$, which is directed into the page ($+z$ direction), lies parallel to the cathode plane.

  \[ \mathbf{B} \perp \mathbf{E} \]

Only **electrons** will be considered because as we have already seen, their dynamical behavior controls glow-discharge processes.
Electron Motion in Combined Electric Field

\( \vec{B} \) and \( \vec{E} \) are parallel

\[
\text{Lorentz force} \quad \vec{F} = \frac{m\vec{v}}{dt} = -q(\vec{E} + \vec{v} \times \vec{B})
\]

\( \vec{v} \), normal to the target surface

\( \vec{v} \), at an angle \( \theta \) to \( \vec{B} \)

\( \vec{E} = 0 \)

\( q \cdot v \sin \theta \cdot B = \frac{m(v \sin \theta)^2}{r} \)

\[\rightarrow r = \frac{mv \sin \theta}{qB}\]

Helical motion with constant radius occurs, but the pitch of the helix lengthens with time.

Clearly, magnetic fields prolong the electron residence time in the discharge and enhance the probability of ion collisions.
Electron Motion in Combined Electric Field

\( \vec{B} \) and \( \vec{E} \) are perpendicular

Electrons emitted normally from the cathode ideally do not even reach the anode but are trapped near the electrode where they execute a periodic hopping motion over its surface.

\[
\begin{align*}
\frac{m_e}{d^t} \frac{d^2 x}{d^t} &= qB \frac{dy}{dt} \\
\frac{m_e}{d^t} \frac{d^2 y}{d^t} &= qE - qB \frac{dx}{dt} \\
\frac{m_e}{d^t} \frac{d^2 z}{d^t} &= 0
\end{align*}
\]
\[ m_e \frac{d^2 y}{dt^2} = q\xi - qB \frac{dx}{dt} \]

\[ m_e \frac{d^2 x}{dt^2} = qB \frac{dy}{dt} \]
\[ m_e \frac{d^2 y}{dt^2} = q\mathcal{E} - qB \frac{dx}{dt} \]

\[ m_e \frac{d^2 x}{dt^2} = qB \frac{dy}{dt} \]
Reflexions and adsorptions give rise to secondary electrons, which can further impinge on the cathode surface and contribute to the overall emission. The secondary electron yield is a critical parameter in cathode-ray tube operation. For a given cathode material and cathode temperature, the yield can be calculated using empirical relationships based on material properties and thermal conditions. These relationships account for factors such as the primary electron energy, the atomic number of the cathode material, and the temperature of the cathode surface. By understanding and optimizing these factors, engineers can achieve the desired characteristics for cathode-ray tubes, leading to high-quality images with clear and distinct features.
Electron Motion in Glow Discharge Plasma

\[ \vec{B} \text{ and } \vec{E} \text{ are perpendicular} \]

- Electrons repeatedly return to the cathode at time intervals of \( \pi/\omega_c \).
- Electron motion is strictly confined to the cathode dark space where both fields are present.
- However, if electrons stray into the negative glow region where \( \vec{E} \) is small, they describe a circular orbit before collisions may drive them either back into the dark space or forward toward the anode.

Confinement in crossed fields prolongs the electron lifetime over and above that in parallel fields, enhancing the ionizing efficiency near the cathode. A denser plasma and larger discharge current result.
- These effects are very widely capitalized upon in magnetron-sputtering processes.
Recall

\[ \frac{dV}{dx} = -E \]

\[ \frac{dE}{dx} = \frac{1}{\varepsilon} q(N^+ - N^-) \]

\[ N^+ - N^- \]

Figure 4-3  Structure of a DC glow discharge with corresponding potential, electric field, charge, and current distributions.

One can see that where there is not a glowing discharge, we either have a greater potential drop or a very low pressure.
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4.3.5 Collective Charge Effects

4.3.5.1 The Debye Length

4.3.5.2 Electron Plasma Frequency

4.3.5.3 Plasma Criteria
Electron Density around an Positive Ion

• The behavior of plasmas derive largely from the Coulombic interactions among the charged species within them.
• There is radial electric potential $V(r)$ around an isolated positive ion.
• This ion repels other ions and attracts a cloud of electrons with a density given by

$$n_e(r) = n_i \cdot e^{\frac{qV(r)}{k_BT}}$$

• The Boltzmann factor reflects the probability that electrons will acquire the energy needed to establish the electric potential at temperature $T$.
• Because $n_e$ cannot deviate much from its average value (which is equal to the ion density $n_i$), $V$ must be small.

$$\therefore n_e \cong n_i \quad \therefore V \cong 0 \quad n_e(r) = n_i \cdot e^{\frac{qV(r)}{k_BT}} = n_i(1 + \frac{qV(r)}{k_BT})$$
Radial Electric Potential around an Isolated Positive Ion

• Furthermore, $V(r)$ must satisfy Poisson’s equation, which in spherical coordinates takes the form,

\[
\frac{1}{r^2} \left[ \left( \frac{d}{dr} \left[ r^2 \frac{dV(r)}{dr} \right] \right) \right] = -\frac{q(n_i - n_e)}{\varepsilon_o} = \frac{n_i q^2 V(r)}{\varepsilon_o k_B T}
\]

Poisson equation

\[
V(r) = \frac{q}{r} \exp\left( -\frac{r}{\lambda_D} \right)
\]

\[
\lambda_D = \sqrt{\frac{\varepsilon_o k_B T}{n_i q^2}}
\]
Debye Length

Debye length \( \lambda_D = \sqrt{\frac{\varepsilon_o k_B T}{n_i q^2}} \)

\[ V(r) = \frac{q}{r} \exp\left(-\frac{r}{\lambda_D}\right) \]

Debye length is a measure of the size of the mobile electron cloud required to reduce \( V \) to 0.37 (i.e., \( 1/e=1/2.71828 \)) of its initial value.

\[ n_i = 10^{10} \text{ cm}^{-3} \]
\[ k_B T = 2 \text{ eV} \]

\[ \rightarrow \quad \lambda_D = 1 \times 10^{-2} \text{ cm} \]

Outside of a sphere of radius \( \lambda_D \) there is effectively no interaction between the ion and the rest of the plasma.

In the case of an inserted electrode, \( \lambda_D \) is a measure of the plasma sheath dimension.
Debye Length for a Charged Planar Electrode

• The same sort of calculation can be performed for a charged planar electrode immersed in the plasma.
• Poisson’s equation in one dimension ($x$) then yields a solution for the potential $V(x)$ that essentially varies as $\exp(-x/\lambda_D)$.
• In the case of an inserted electrode, $\lambda_D$ is a measure of the plasma sheath dimension.
4.3.5 Collective Charge Effects
   4.3.5.1 The Debye Length
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   4.3.5.3 Plasma Criteria
Plasma Oscillation

• By evaluating its response to a perturbation, the ability of a plasma to protect its charge neutrality can be assessed.
• Consider that an external electric field is suddenly turned on, displacing plasma electrons over some length.
• If it is just as suddenly turned off, the electron displacement induces a field that pulls the electrons back to their original position.
• But the inertia of the electrons will cause them to overshoot the mark and harmonically oscillate about the equilibrium sit.
Electron Plasma Frequency

When charged particles are separated by a short distance $x$ the polarization, $P = qnx$, where $n = n_+ = n_-$ is the number density of the particles.

$$P = qnx$$

The restoring electric field $E$ is given by $-P / \varepsilon_o$ so

$$E = -\frac{P}{\varepsilon_o}$$

$$F = m \frac{d^2 x}{dt^2} = qE = -q \frac{P}{\varepsilon_o} = -\frac{nq^2 x}{\varepsilon_o}$$

Simple Harmonic Motion

$$F = -kx \rightarrow k = \frac{nq^2}{\varepsilon_o}$$

$$f = \frac{1}{2\pi} \sqrt{\frac{k}{m}}$$

$$\omega = \sqrt{\frac{k}{m}} = \sqrt{\frac{nq^2}{m\varepsilon_o}}$$

Electron plasma frequency $\omega_p$, given by

$$\omega_p^2 = \frac{ne^2}{m_e \varepsilon_o}$$
Electron Plasma Frequency

\[ \omega_e = \sqrt{\frac{q^2 n_e}{m_e \varepsilon_o}} = 8.98 \times 10^3 \sqrt{n_e} \text{ Hz} \]

(n_e : number density of electrons)

\[ \lambda_D = \sqrt{\frac{\varepsilon_o k_B T}{n_i q_e^2}} \quad \rightarrow \quad \omega_e \lambda_D = \sqrt{\frac{q^2 n_e}{m_e \varepsilon_o}} \sqrt{\frac{\varepsilon_o k_B T}{n_i q_e^2}} = \sqrt{\frac{k_B T}{m_e}} = v_e \]

The product of \( \lambda_D \) and \( \omega_e \) is essentially equal to the electron velocity.

If the frequency is less than \( \omega_e \) the dielectric constant is high and the plasma appears *opaque* to such radiation.

On the other hand the plasma becomes *transparent* to radiation at frequencies greater than \( \omega_e \) where the dielectric constant drops.

For \( n_e = 10^{10} \text{ cm}^{-3}, \omega_e = 9 \times 10^8 \text{ Hz} \), a frequency much larger than that typically used in AC (RF) plasmas.
Relative permittivity is also commonly known as dielectric constant.

Relative permittivity \( \varepsilon_r(\omega) = \frac{\varepsilon(\omega)}{\varepsilon_0} \)

Relative permittivity is the ratio of the capacitance of a capacitor using that material as a dielectric, compared with a similar capacitor that has vacuum as its dielectric.

\[
\varepsilon_r = \frac{C}{C_0}
\]

\[
C = \varepsilon \frac{A}{d} = \varepsilon_r \varepsilon_0 \frac{A}{d} = \varepsilon_r C_0
\]

\[
C \equiv \frac{Q}{V}
\]
4.3.5 Collective Charge Effects
4.3.5.1 The Debye Length
4.3.5.2 Electron Plasma Frequency
4.3.5.3 Plasma Criteria
Ionized gases can be characterized as plasmas if they meet three criteria:

1. **System dimension >> Debye length**
   - $D >> \lambda_D$
   - Only in this way the quasineutrality of the bulk of the plasma be ensured.

2. **Shielding electrons drawn into the Debye sphere must be large.**
   - $N_D >> 1$ (at least)
   - $N_D = 4\pi \lambda_D^3 n_e/3$
   - $N_D \sim 4 \times 10^4$

3. **Electrons should interact more strongly with each other than with the neutral gas.**
   - Under this conditions, particle motion in the plasma will be controlled by electromagnetic forces rather than by gas fluid dynamics.

$$\lambda_D = \sqrt{\frac{\varepsilon_0 k_B T}{n_i q^2}} \quad n_i = n_e = 10^{10} \text{ cm}^{-3} \quad k_B T = 2 eV \quad \rightarrow \quad \lambda_D = 1 \times 10^{-2} \text{ cm}$$

$$N_D = \frac{4\pi \lambda_D^3 n_e}{3} \sim 4 \times 10^4$$
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AC Effects in Plasmas

- How plasmas are sustained can be appreciated by analyzing kinetic behavior of electrons.
- In their to and fro motion in the field, electrons would absorb and gain sufficient energy to cause enhanced ionization of neutrals.
- When the electrons undergo inelastic collisions their motion is randomized and power is effectively absorbed from the RF source.
Field Strength Required to Ionize Gas

Assuming no collisions of electrons with neutrals, \( m_e \frac{d^2 x}{dt^2} = -q \mathcal{E}_o \sin \omega t \)

\[
x_o = \frac{q \mathcal{E}_o}{m_e \omega^2} \quad \text{Maximum electron displacement amplitude}
\]

\[
E_o = \frac{1}{2} (q \mathcal{E}_o) \cdot x_o = \frac{(q \mathcal{E}_o)^2}{2m_e \omega^2} \quad \text{Ionization energy}
\]

\[ E_{o,(Ar)} = 15.7 \text{eV} \]

\[ f = 13.56 \text{MHz} \quad (\omega = 2\pi \times 13.56 \times 10^6 \text{Hz}) \]

\[ \rightarrow \quad \mathcal{E}_o = 11.5 \text{ V/cm} \]

\( \mathcal{E}_o = 11.5 \text{ V/cm} \) is an easily attainable field in typical plasma reactors. No power is absorbed in the collisionless harmonic motion of electrons, however.

For electrons undergo inelastic collisions,

Electron motion is randomized and power is effectively absorbed from the RF source. Even smaller values of \( \mathcal{E}_o \) can produce ionization if, after electron-gas collisions, the reversal in electron velocity coincides with the changing electric-field direction. Through such effects RF discharges are more efficient than their DC counterparts in promoting ionization.
Electrodeless Reactors

Interestingly, provided the frequency is high enough, reactors can be built without interior plate electrodes.

- A coil wrapped around a tubular reactor can inductively couple power to the gas inside ionizing it.

- So, too, can capacitor plates on the outside; in such a case we speak of capacitive coupling.

Such electrodeless reactors have been used for etching films. However, for the deposition of films by RF sputtering, internal cathode targets are required.
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Immersion of a floating electrode into a plasma causes it to charge negatively because of the disparity (in mass, velocity, and energy) between electrons and ions. The electrode receives a higher flux of electrons, compared to ions, and charges negatively. The resulting negative potential reduces the electron flux, such that it again equals the ion flux.
1. As a consequence, we can expect that both anode and cathode surfaces will be at a negative floating potential ($V_f$) relative to the plasma potential ($V_p$).

2. In essence a Debye-like, positive space charge layer shields the negative surface; we now speak of a plasma sheath of potential $V_s$ that envelops the electrode.

- Lower electron density in the sheath means less ionization and excitation of neutrals.
- Less luminosity there.
- Large electric fields are restricted to the sheath regions.
- It is at the sheath-plasma interface that ions begin to accelerate on their way to the target during sputtering.
- The plasma is typically some ~ 15 volts positive with respect to the anode.

$$V_s = V_p - V_f$$
Sheaths Potential Barrier

It is not difficult to quantitatively sketch the magnitude of the potential energy barrier $q(V_p - V_f)$ electrons face in moving from the plasma to the cathode surface through the sheath.

The number of electrons ($n_e^*$) that can gain enough energy to surmount this barrier is given by

$$\frac{n_e^*}{n_e} = \exp - \frac{q(V_p - V_f)}{k_B T}$$

Considering the electrical flux balance between electrons and ions near the cathode, it gives

$$\frac{n_e^*}{n_e} = \frac{2.3m_e}{m_i}$$

(Derivation detail not given.)

$$\Rightarrow V_p - V_f = \frac{k_B T e}{2q} \ln\left(\frac{m_i}{2.3m_e}\right) = e.g., \sim 10eV$$

since $m_i$ is 3-4 orders of magnitude higher than $m_e$. 

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Ion Current Flow

Ion current flow through the cathode sheath is an important issue because all thin-film processing in plasmas depends on it. In this regard the interesting question arises as to whether ion motion in the sheath occurs in a “free-fall,” collisionless manner, or through “mobility limited” motion involving repeated collisions with other gas species.

“Free fall”, collisionless manner

\[
j = \frac{4\varepsilon_0}{9d_s^2} \sqrt{\frac{2q}{m}} \cdot V^{3/2} \quad \text{Free fall or space charge limited}
\]

“Mobility limited”, repeated collision

\[
j = \frac{9\varepsilon_0}{8d_s^3} \cdot V^2 \quad \text{Mobility limited}
\]

\(V\): voltage applied across a sheath thickness
\(d_s\): sheath thickness

It turns out that free fall model, also known as the Child-Langmuir equation, better describes the measured cathode-current characteristics. This is certainly true at low pressures where few collisions are likely.
The sheath dark space is sometimes visible with the unaided eye and is therefore considerably larger than calculated **Debye lengths** of 100 $\mu m$.

This means that a large planar surface behaves differently from a point charge when both are immersed in plasmas. Instead of electrons shielding a point charge, bipolar diffusion of both electrons and ions is required to shield an electrode, and this physically broadens the sheath dimensions.

A useful formula suggests that the relation between $d_s$ and $\lambda_D$ is


$$d_s \sim \left[ \frac{q(V_p - V_f)}{k_B T_e} \right]^a \cdot \lambda_D$$

$$\frac{2}{3} \text{ (higher pressures)} < a < \frac{3}{4} \text{ (lower pressures)}$$

$$d_s \sim n \times 10 \cdot \lambda_D$$