Low Temperature Plasmas

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Day 9....on Zoom!

How did plasma physics find me?

- Originally from northern Wisconsin
 - Like to build things -> engineering school
- University of Wisconsin
 - Nuclear Engineering/Engineering Physics (BS 2006, MS 2008, PhD 2010)
 - Undergrad RA in Prof. Hershkowitz's lab
 - Caught the plasma bug, never looked back
- Postdoctoral training
 - University of New Hampshire (2010-2012)
 - Los Alamos National Laboratory (2012-2013)
- University of Iowa Physics and Astronomy



Prof. Noah Hershkowitz Irving Langmuir Professor (emeritus) University of Wisconsin

Why plasma physics and engineering?

- 1. The thrill of "finding things out"
- 2. Can be both fundamental and practical
 - Fundamental questions: matter in extreme conditions, nonlinear dynamics, complexity, self-organization, etc., etc.
 - Translational: Fundamental discovery often leads to new devices on a short timescale
 - Contributes to solving pressing problems of society
 - This is especially true in <u>Low Temperature Plasmas</u>



Outline of this talk

- What is a low temperature plasma?
- What are they used for?
- Historical context
- Basic properties
 - Plasma potential
 - Sheaths
 - Plasma chemistry



What is a low temperature plasma?

Low temperature? I thought plasmas were hot

- Low temperature plasmas (LTP) are roughly defined by:
 - Ion temperature: $T_i \sim 0.03 \text{eV}$ (near room temp)
 - Electron temperature: $T_e \sim 1 100 \text{eV}$ (few-to-several eV)
- Highly non-equilibrium
 - "Temperature" refers to a mean kinetic energy, rather than a thermodynamic temperature
- Plasma density can vary widely: $n \sim 10^6 10^{16} \text{ cm}^{-3}$
- Ionization fraction is usually low: $\frac{n_p}{n_n} < 1\%$

How can electron and ion "temperatures" be so different?

Ions are much more massive than electrons

- Electron mass: $m_e = 9.1 \times 10^{-31}$ kg
- Proton mass: $m_i = 1.7 \times 10^{-27} \text{kg}$
- Ratio: $\frac{m_i}{m_e} \approx 1836$
- About the same as:
- Ping pong ball: $m_{pp} = 2.7$ g
- Bowling ball: $m_{bb} = 5 \text{ kg}$
- Ratio: $\frac{m_{bb}}{m_{pp}} \approx 1850$







Electrons move much faster than ions

• Thermal speed: $v_{T\alpha} = \sqrt{\frac{2k_B T_{\alpha}}{m_{\alpha}}}$

• Ratio:
$$\frac{v_{Te}}{v_{Ti}} = \sqrt{\frac{m_i T_e}{m_e T_i}} \approx 250 \text{ (uses } T_e = 1 \text{eV)}$$

About the same as

- A person walking: 3 miles/hr
- The speed of sound: 760 miles/hr

• Ratio:
$$\frac{v_s}{v_w} \approx \frac{760}{3} \approx 250$$







Equilibration rate depends on the mass ratio

• Reference collision rate (electron-electron)

$$v_o^e(s^{-1}) \approx \frac{5 \times 10^{-11} n(m^{-3})}{[T(eV)]^{3/2}}$$

- Mean free path: $\lambda_{\varepsilon}^{\alpha\beta} = \frac{\nu_{T\alpha}}{\nu_{\varepsilon}^{\alpha\beta}}$
- Time (or distance) to come to same temperature

$$\begin{split} \nu_{\varepsilon}^{ee} &\approx \nu_{o}^{e} \\ \nu_{\varepsilon}^{ei} &\approx \frac{m_{e}}{m_{i}} \nu_{o}^{e} \ll \nu_{\varepsilon}^{ee} \\ \nu_{\varepsilon}^{ii} &\approx \sqrt{\frac{m_{e}}{m_{i}}} \left(\frac{T_{e}}{T_{i}}\right)^{3/2} \nu_{o}^{e} &\approx \nu_{\varepsilon}^{ee} \end{split}$$

$$\lambda_{\varepsilon}^{ee}(m) \approx 1 \times 10^{16} \frac{[T_e(eV)]^2}{n(m^{-3})}$$
$$\lambda_{\varepsilon}^{ei} \approx \frac{m_i}{m_e} \lambda_{\varepsilon}^{ee} \gg \lambda_{\varepsilon}^{ee}$$
$$\lambda_{\varepsilon}^{ii} \approx \left(\frac{T_i}{T_e}\right)^2 \lambda_{\varepsilon}^{ee} \ll \lambda_{\varepsilon}^{ee}$$

Electrons escape the plasma before they come into equilibrium with ions

- Example: $T_e = 1eV$, $T_i = 0.03eV$, n = 1×10¹⁶ m^{-3} , $L \approx 1$ m, protons
- Mean free paths:

 $\begin{array}{l} \lambda_{\varepsilon}^{ee} \approx 1 \text{ m} \longrightarrow \text{Marginal e-e equilibration} \\ \lambda_{\varepsilon}^{ei} \approx 2000 \text{ m} \longrightarrow \text{No e-i equilibration} \\ \lambda_{\varepsilon}^{ii} \approx 0.001 \text{m} \longrightarrow \text{Excellent i-i equilibration} \end{array}$

- Ions equilibrate with the neutrals
 - Similar to ion-ion mean free path
 - Details depend on pressure



$$\lambda_{ei} \gg L$$

$$\lambda_{ee} \sim L$$

$$\lambda_{ii} \sim \lambda_{in} \ll L$$

LTPs are (relatively) easy to make

- Power source couples energy to electrons
 - Many ways to do this (a few examples to come)
- Energetic electrons partially ionize gas
 - Ionization energies ~10s eV
- A simple example is an incandescent light bulb
 - Electrons emitted from a hot filament
- Unlike fusion plasmas, <u>LTPs are typically not</u> <u>"confined"</u> by a magnetic field
 - Electrons and ions are lost to boundaries



Image from: https://www.1000bulbs.com



dc filament-driven discharges

- Heat material that emits electrons when hot
 - Thermionic emission
 - Filament
- dc bias the filament with respect to the plasma
 - Electrons gain energy from the electric field between the filament and plasma
- Energetic electrons ionize gas





CHAMBER/ANODE

dc electric field-driven discharges

- Electric field accelerates electrons to high energy
 - Electrons ionize neutrals
- Examples
 - Arcs and sparks
 - Glow discharges







Image from:

https://commons.wikimedia.org/wiki/File:Glow_discharge_regions.jpg

Image from: https://upload.wikimedia.org/wikipedia/commons/e/eb/Staccoto_Lightning.jpg

ac capacitively coupled discharges

- Electrodes biased with ac voltage
 - MHz frequency <u>electrostatically</u> couples power to electrons
 - Advantageous because electric fields are confined to edges (sheaths)





Figure from: Pattyn, Kovacevic, Strunskus, Lecas and Bernt, Scientific Reports 6, 10886 (2019).

ac inductively coupled discharges

- Antenna biased with ac voltage
 - 10s of MHz to couple with electrons
- Energy coupled by <u>electromagnetic</u> <u>induction</u>
 - ac current in the coils induces an ac magnetic field inside the tube
 - produces an electromotive force (electric field) inside the plasma
 - Energetic electrons ionize neutrals



Image from: https://www.trumpf.com



Low temperature plasmas are tremendously useful



Utilize directed energy of charged particles

- Electric fields accelerate ions (or electrons) in a preferred direction
- Directed kinetic energy is used to do useful things
 - Etching semiconductors
 - >\$1 T industry!
 - Surface modification of materials
 - Plasma-based propulsion
 - Ion and electron sources
 - Sources of fusion neutrons
 - neutron imaging
 - producing medical isotopes
 - (not for net energy production)



www.sentech.com



Non-equilibrium (high temperature electrons) enables unique chemistry

- "Normal" chemistry only accesses reactions near room temperature
 - New reactions at eV e⁻ energy
 - Plasma catalysis
- Many applications
 - Materials processing
 - Climate
 - Chemical production
 - Medicine
 - Energy
 - Agriculture



Figure from: Zhang et al, "Plasma activation of methane for hydrogen production in a N2 rotating gliding arc warm plasma: A chemical kinetics study," Chemical Engineering Journal 345, 67 (2018).

Science Challenges in Low-Temperature Plasma Science and Engineering: Enabling a Future Based on Electricity Through Non-Equilibrium Plasma Chemistry



https://mipse.umich.edu/nsfworkshop/docs/NSF_LTP_Workshop_Report_2017_04_01_2side_print.pdf

Historical context



Joseph J. Thomson (1856-1940)

- Cavendish Professor of Physics, Cambridge
- Discovered
 - electrons, isotopes, Thomson scattering, plum pudding model, mass spectrometer, waveguides
- Renowned educator
 - Advised 6 physics Nobel laureates and 2 chemistry Nobel laureates
 - Ex: Rutherford, Bohr, Born, Oppenheimer, Bragg





(1906) for work on the conduction of electricity in gasses

Thomson discovered the electron

- Needed high vacuum
 otherwise a plasma would form!
- Cathode rays were influenced by electric and magnetic fields
 - Measured charge-to-mass ratio
- Understanding collisions was essential
 - Showed size is 1000x smaller than the atom (first subatomic particle)
- Plum pudding model of the atom
 - corpuscles emanating in a sea of positive charge

Crooks tube with high vacuum



From: wikipedia.org

Irving Langmuir (1881-1957)

- Industrial physicist at General Electric
- Founded
 - Plasma physics
 - Surface chemistry
 - Atmospheric control (cloud seeding)
- Invented gas-filled incandescent lamp, Langmuir probe diagnostic, Langmuir (plasma) wave, hydrogen welding





1932: For his discoveries and inventions in surface chemistry

Langmuir named "plasma" and "sheath"

(from I. Langmuir, "Oscillations in Ionized Gases," Proc. Nat. Acad. Sci. 14, 627, August 1928)

"Except near the electrodes, where there are *sheaths* containing very few electrons, the ionized gas contains ions and electrons in about equal numbers, so that the resultant space charge is very small. We shall use the name *plasma* to describe this region containing balanced charges of ions and electrons."



Hull and Langmuir, Proc. Nat. Acad. Sci. 51, 218 (1929)

Langmuir named "plasma" and "sheath"

staffs—who will not become Chief Scientists or Permanent Secretaries with a pay and career structure which matches that of those of their colleagues whose job weight, background and qualifications is unquestionably in no way superior to their own. That is not the case now; we are quite determined that it shall become so.

All this is far removed from your suggestion that we may become "entirely lost in detailed quibbles", or that we are taking an insufficiently radical view of what is needed. I should have thought that our aims are precisely those which would result in scientists assuming their rightful position in the Service. As you have recognized, it will be a sorry day for science and scientists generally if we fail. We do not intend to fail; there is too much at stake.

Yours faithfully,

CYRIL COOPER

Deputy General Secretary

The Institution of Professional Civil Servants, Northumberland Street, London WC2

History of "Plasmas"

SIR,—There has been speculation as to the basis for Irving Langmuir's use of the word "plasma" to describe a particular region of an electrical discharge in a gas. I was working for Langmuir when he made this innovation about 1927, and, much later, described it in a letter to a friend at the General Electric Research and Development Center, dated April 20, 1967. The pertinent extract from this letter follows.

"Meanwhile word had come down from on high that we had better do something about mercury-arc rectifiers and converters, which were the big new things in the electrical engineering world, although at that time they were all in glass bulbs. So Langmuir began to study mercury vapor discharges. He shortly invented his probe, I did the experimental work and most of the mathematics, and we soon accumulated a lot of data about ion densities and velocity distributions in the mercury arc columns, in the positive columns of Geissler tubes, and in gasfilled thermionic tubes.

"We noticed the similarity of the discharge structures they revealed. Langmuir pointed out the importance and probable wide bearing of this fact. We struggled to find a name for it. For all members of the team realized that the credit for a discovery goes not to the man who makes it, but to the man who names it. Witness the name of our continent. We tossed around names like 'uniform discharge', 'homogeneous discharge', 'equilibrum discharge'; and for the dark or light regions surrounding

electrodes, names like 'auras', 'haloes', and so forth. But one day Langmuir came in triumphantly and said he had it. He pointed out that the 'equilibrium' part of the discharge acted as a sort of sub-stratum carrying particles of special kinds, like high-velocity electrons from thermionic filaments, molecules and ions of gas impurities. This reminds him of of the way blood plasma carries around red and white corpuscles and germs. So he proposed to call our 'uniform discharge' a 'plasma'. Of course we all agreed.

"But then we were in for it. For a long time we were pestered by requests from medical journals for reprints of our articles. This happens to me this day. The scientific world of physics and chemistry looked askance at this uncouth word and were slow to accept it in their The engineering world vocabulary. treated it as a GE trade name. Then all of a sudden, long after I had left the laboratory, to my pleased surprise, everybody started to talk about plasmas. This happened not long before they became thermonuclear and so government subsidized. That finally put the seal of respectability on plasmas."

Yours faithfully,

HAROLD M. MOTT-SMITH

6909 SW Freeway, Texas 77036

Basic Properties



Plasmas are surrounded by sheaths

- <u>Sheath</u>: A narrow region of strong electric field that acts to balance electron and ion currents lost from a plasma
- Recall: electrons move faster than ions

$$\frac{v_{Te}}{v_{Ti}} = \sqrt{\frac{m_i T_e}{m_e T_i}} \approx 250 \text{ (uses } T_e = 1\text{eV)}$$

- Electrons quickly charge the boundary surface
- Sheaths are an electrostatic barrier that forms naturally as ions are attracted to the electron surface charge





Sheaths are thin (almost like a skin)

• Polarization shields electric fields in plasmas

• Debye length:
$$\lambda_{De} = \sqrt{\frac{\epsilon_0 T_e}{en}} = 740 \sqrt{\frac{T_e(eV)}{n(cm^{-3})}} \text{ cm}$$

• For our example:
$$T_e = 1eV$$
, $n = 1 \times 10^{10} cm^{-3}$:

$$\lambda_{De} \approx 7 \times 10^{-5} \mathrm{m}$$
 (tiny!)

• More complete analysis of sheath thickness (Child-Langmuir):

$$\frac{l_s}{\lambda_D} = \frac{\sqrt{2}}{3} \left(\frac{2e\Delta\phi}{T_e}\right)^3$$

- Plasma potential: $\Delta \phi$
 - Determined by balancing electron and ion currents to boundary

David Bohm (1917-1992)

- Leading theoretical physicist of the 20th century
 - Hidden variables theory of quantum mechanics, Aharonov-Bohm effect, neuropsychology, philosophy
 - In plasma: Bohm diffusion, Bohm criterion, plasmon, random-phase approximation
- His war work was in plasma
 - Refused security clearance
 - Thomson -> Oppenhiemer -> Bohm





Fellow of the Royal Society Expert in theoretical physics, philosophy and neuropsychology

(photo credit: www.wikipedia.org)

Bohm criterion and the presheath

• Ions must flow into the sheath supersonically

$$V \geq c_s = \sqrt{T_e/m_i}$$





Bohm, in Characteristics of Electrical Discharges in Magnetic Fields, 1949

Plasma potential is determined by balancing electron and ion currents

• Ions are lost at the Bohm flux:

$$\Gamma_i \approx 0.6 en \sqrt{\frac{T_e}{m_i}}$$

- The 0.6 is from the presheath density drop
- Electrons are lost a thermal flux, reduced by the sheath:

$$\Gamma_e \approx en \sqrt{\frac{T_e}{2\pi m_e}} e^{-\Delta \phi/T_e}$$

• Setting these equal gives the <u>plasma potential</u>:

$$\Delta \phi = \frac{T_e}{e} \ln \left(\sqrt{\frac{\pi m_i}{8m_e}} \right) \approx 3 \mathrm{V}$$

Summary of sheath properties

- 2 length scales
 - Sheath $\sim \lambda_D$
 - Presheath $\sim \lambda_{in} \gg \lambda_D$
- Child-Langmuir law $\frac{s}{\lambda_D} = 0.79 \left(\frac{e\Delta\phi}{T_e}\right)^{3/4}$
- Bohm criterion

$$V_i \ge c_s = \sqrt{T_e/m_i}$$

- Presheath potential drop

 $e\Delta\phi_{ps} \ge T_e/2$



Plasma interaction with electric fields, such as sheaths, are the basis for many technologies

- Manufacturing microelectronics
 - Directed ion energy enables etching of large aspect ratio trenches
 - Enabled Moore's law
- Chemistry at the surface is also important
 - Reactive ion etching



Figure from: Samukawa, et al "2012 Plasma Roadmap," J. Phys. D 45, 253001 (2012).

High Aspect Ratio



Figure from: http://www.appliedmaterials.com/

Sheaths also influence the chemistry

- Transfer of plasma-generated reactive species to materials
 - Happens through a sheath
- The sheath influences the electron energy distribution function (EEDF) in the plasma
 - The EEDF controls the rate of chemical reactions
 - Modeling the EEDF is a complex plasma-boundary interaction problem, mediated by the sheath



Figure from: Samukawa, et al "2012 Plasma Roadmap," J. Phys. D 45, 253001 (2012).



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Plasmas enable more of our modern way of life than you might realize



01—Plasma TV	09—Plasma-aided combustion
02—Plasma-coated jet turbine blades	10—Plasma muffler
03—Plasma-manufactured LEDs in panel	11—Plasma ozone water purifica
04—Diamondlike plasma CVD	12—Plasma-deposited LCD scree
eyeglass coating	13—Plasma-deposited silicon fo
05—Plasma ion-implanted artificial hip	solar cells
06—Plasma laser-cut cloth	14—Plasma-processed microele
07—Plasma HID headlamps	15—Plasma-sterilization in pharmaceutical production
08—Plasma-produced H in fuel cell	

bustion 16—Plasma-treated polymers 17—Plasma-treated textiles er purification 18—Plasma-treated heart stent LCD screen 19—Plasma-deposited diffusion barriers silicon for for containers 20—Plasma-sputtered window glazing microelectronics 21—Compact fluorescent plasma lamp n in

Figure from "Plasma 2010" NAS decadal report

Low Temperature Plasma is an exciting area of science and engineering

- Fast-paced
 - Short timeframe from fundamental research to engineering implementation
- Interdisciplinary
 - Source design must be coupled with desired outcome
- Important
 - Enables many modern technologies
 - And you may not have even realized it!



Suggested References

WILEY

Principles of Plasma Discharges and Materials Processing

Second Edition

Michael A. Lieberman and Allan J. Lichtenberg





OPEN ACCESS IOP Publishing J. Phys. D: Appl. Phys. 50 (2017) 323001 (46pp)

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https://doi.org/10.1088/1361-6463/aa76f5

Topical Review

The 2017 Plasma Roadmap: Low temperature plasma science and technology

I Adamovich¹, S D Baalrud², A Bogaerts³, P J Bruggeman⁴, M Cappelli⁵, V Colombo⁶, U Czarnetzki⁷, U Ebert^{5,9}, J G Eden¹⁰, P Favia¹¹, D B Graves¹², S Hamaguchi¹³, G Hieftje¹⁴, M Horl¹⁵, I D Kaganovich¹⁶, U Kortshagen⁴, M J Kushner¹⁷, N J Mason¹⁸, S Mazouffre¹⁹, S Mededovic Thagard²⁰, H-R Metelmann²¹, A Mizuno²², E Moreau²³, A B Murphy²⁴, B A Niemira²⁴, G S Oehrlein²⁶, Z Lj Petrovic²⁷, L C Pitchford²⁸, Y-K Pu²⁹, S Rauf³⁰, O Sakai³¹, S Samukawa³², S Starikovskaia³³, J Tenyson³⁴, K Terashima³⁵, M M Turner³⁶, M C M van de Sanden^{9,37} and A Vardelle³⁸