

Classical Langmuir probe diagnostics: Validity, problems and their resolution

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It will be shown that classical probe diagnostics based on electron and ion parts of the probe characteristics can be in significant error. The reason for errors in plasma parameter evaluation is negligence of limitations in probe diagnostic theories which sometime are not suitable for a real plasma experiment. The main problem in application of the Langmuir probes is the absence of a Maxwellian electron energy distribution function, EEDF (assumed in probe theories) and others non-realistic assumptions. EEDF governs electron and ion current to the probe, and plasma reaction rates. Detailed comparison of plasma parameters, measured with different Langmuir probes, with microwave probes and found as integrals of the measured EEDF are discussed in this presentation. It has been shown that plasma parameters inferred from the ion current to probe can be in error up to order of magnitude. It is explained by different effects of probe-plasma interactions unaccounted in the ion current theories. The main conclusion in this presentation is that the measurement of EEDF following its integration is the only accurate and reliable method of contemporary probe diagnostics.

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Langmuir probe usage

Langmuir probes with spectroscopy were used for a century as the main experimental tools in study of gas discharge plasmas. Many good probe papers and reviews were published since, but the majority of today publications on probe diagnostics are of mediocre and bad quality comparing to those made a half century ago.

There are three main Langmuir probe diagnostic methods:

1. Classical Langmuir probe based on electron part of the probe I/V characteristic
2. Probe diagnostics based on ion part of the probe I/V characteristic (floating single, double and triple probes) now days became classical
3. Measurement of EEDF by differentiation of the probe I/V characteristic

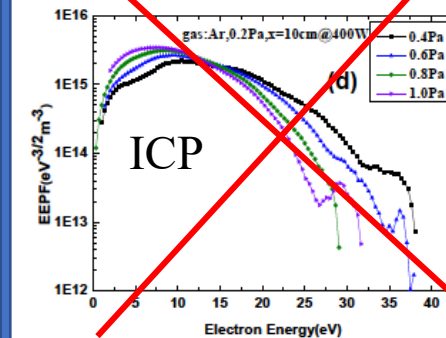
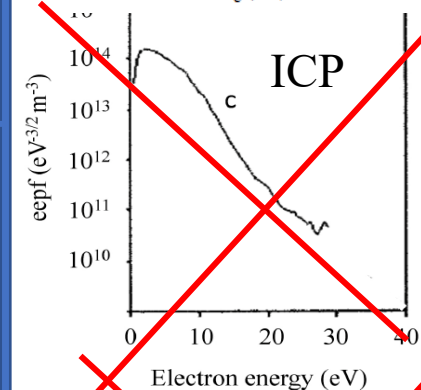
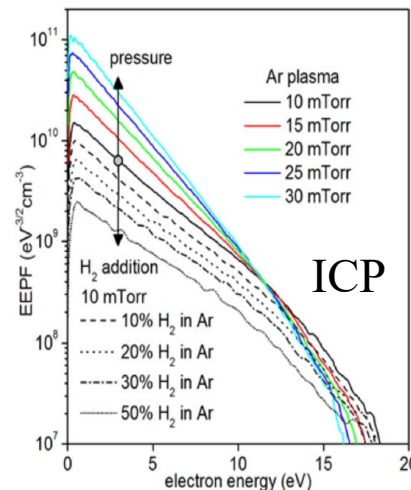
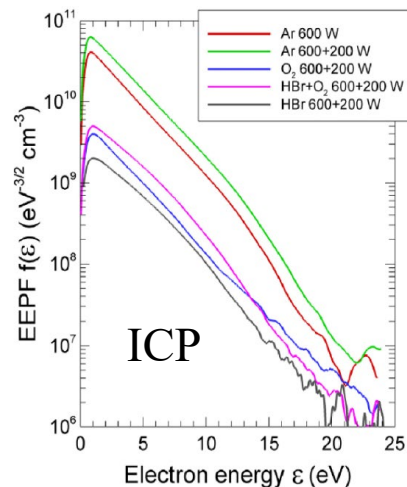
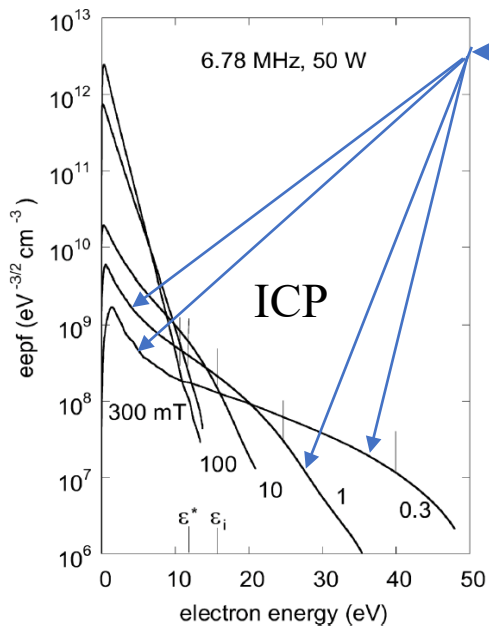
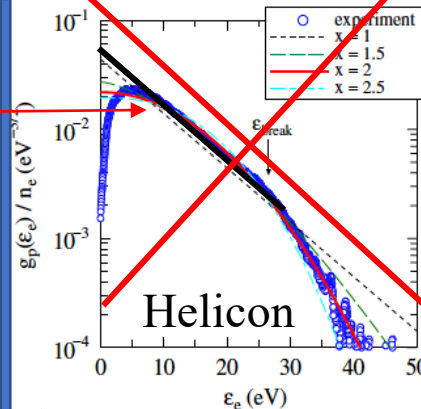
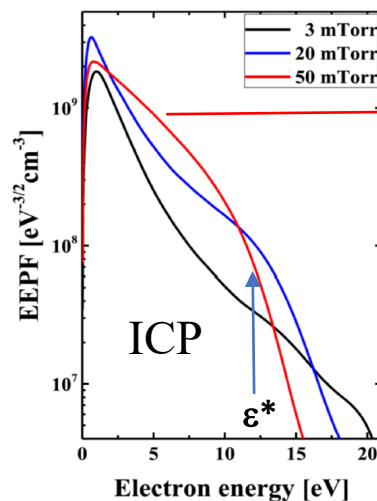
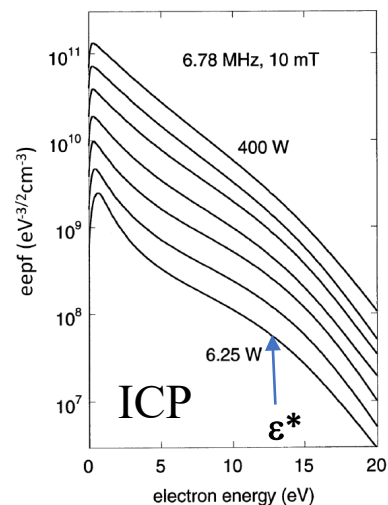
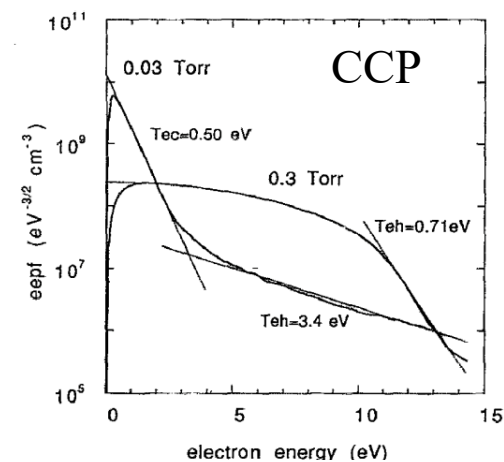
These techniques are essentially different in complexity, obtained information and validity

Basic assumptions in Langmuir probe diagnostics

- Inferred from probe measurement plasma parameters are undisturbed (by the probe) plasma parameters.
- Maxwellian EEDF (isotropic, $F_1 \ll F_0$, $B = 0$, limited E/N).
- No electron collisions in perturbed zone around the probe ($\lambda_e \gg a \cdot \ln[\pi l/4a]$, b , λ_D)
Here **a** and **b** are the probe and its holder radius, **l** is the probe length, λ_e and λ_D are the electron mean free path and Debye length.
- Small probe (no impact on discharge current, electron energy and particle balance ($I_p \ll I_d$, I_{iw} , I_ε). $I_{iw} = S_{ch} e n_w v_s$ and $I_\varepsilon(\varepsilon)$ is the flux of electrons due to their generation in the volume $\approx \lambda_e^3$, or Λ^3 when $\Lambda < \lambda_e$. Here Λ is the characteristic plasma size and λ_e is the the electron energy relaxation length.
- Small probe circuit resistance $R_c \ll R_{pmin} = T_e/eI_{eo}$, otherwise, $V_{psh} < V_p$.
- No large low and high frequency voltage across the probe sheath affecting dc probe I/V characteristic. Probe I/V is non-linear!
- Clean probe surface and constant probe work-function during probe voltage span.

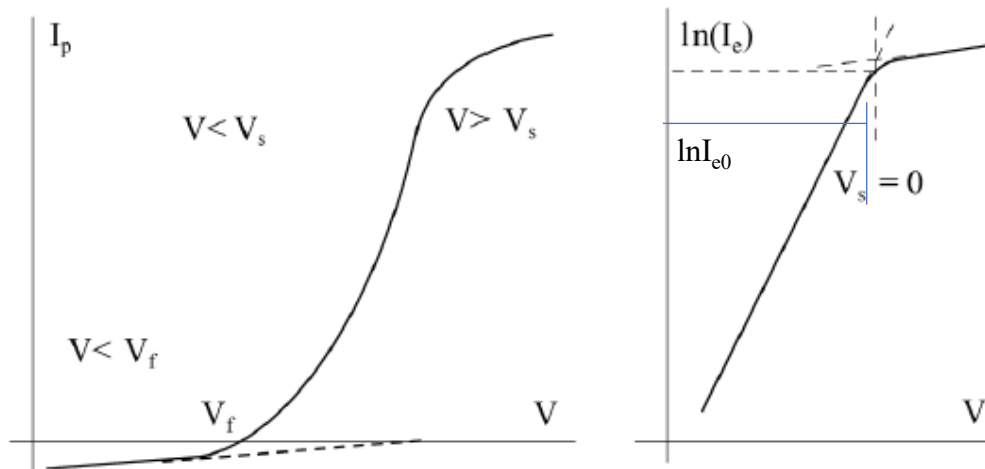
Violation of these conditions is practically invisible in the measured probe I/V characteristics, but can be observed in its second derivative d^2I_p/dV_p^2 .

EEDF in gas discharge plasma is non-Maxwellian in the range of relevant
electron energy roughly defined as $0.2 [\epsilon] \leq \epsilon \leq \epsilon_i + 2[\epsilon]$



In industrial plasma processing reactors

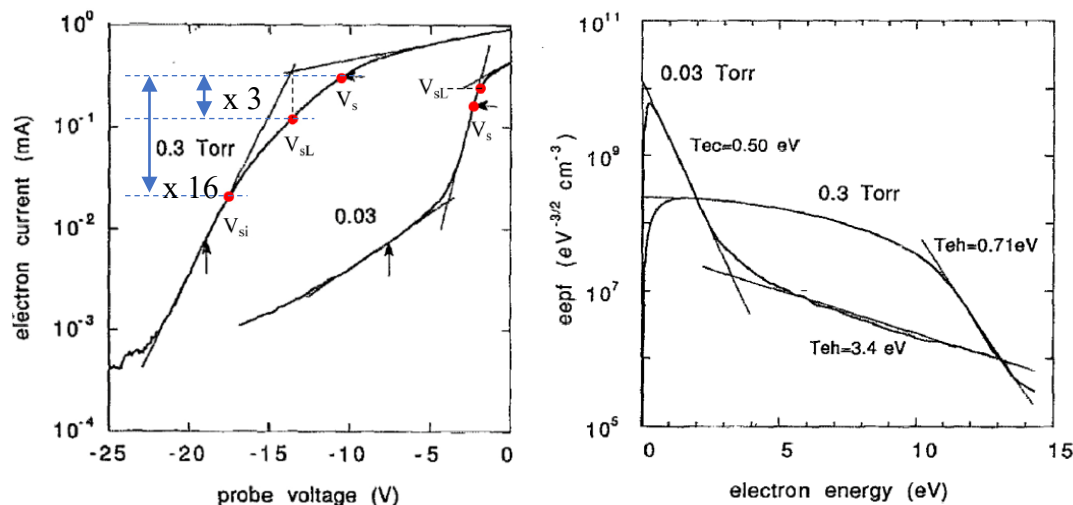
Classical Langmuir probe



Probe characteristics given in textbooks

In practice, for properly designed probes, there is no clearly a distinctive brake point on the $\ln[I_e(V)]$ curve; the transition is relatively smooth. The experimental probe characteristic looks like in textbooks only when the probe is large or/and the plasma density is very high.

The desire to obtain a “looking nice as in textbooks” probe characteristics leads some beginners to use excessively large probes. Such probes create the plasma disturbances and also may cause the voltage applied to the probe to split between the probe sheath and others elements of the probe circuit.

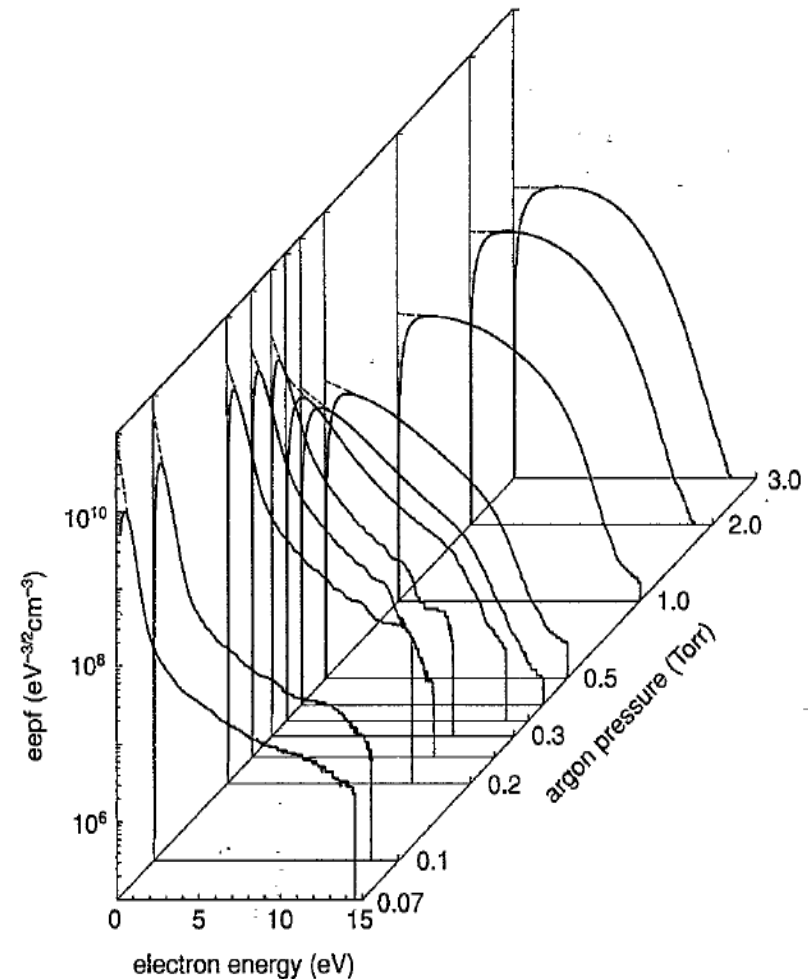


Probe characteristics and EEPF in Ar CCP & 13.56 MHz

Druyvestein-like EEDF is typical in Ramsauer gases, like Ar, Kr, Xe dc and rf plasma at low discharge power (plasma density) when electron-electron collision frequency $\nu_{ee} \propto n_e^{-3/2}$ is small. Therefore, the results of Langmuir probe measurements in such plasmas should be in significant error.

The main problems in the classical Langmuir probe diagnostics (providing that the others requirements are satisfied)

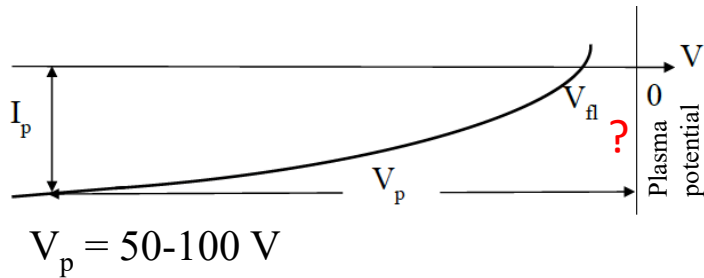
- The difference between bad and good probe characteristics are practically invisible.
- The absence of Maxwellian EEDF in low ($\varepsilon < \varepsilon^*$) and high ($\varepsilon > \varepsilon^*$) electron energy.
- Uncertainty in plasma potential, thus, in electron saturation current and plasma density.
- Uncertainty in approximation of the ion part of the probe characteristic affecting accuracy in evaluation of T_e for fast electrons governing excitation and ionization processes.



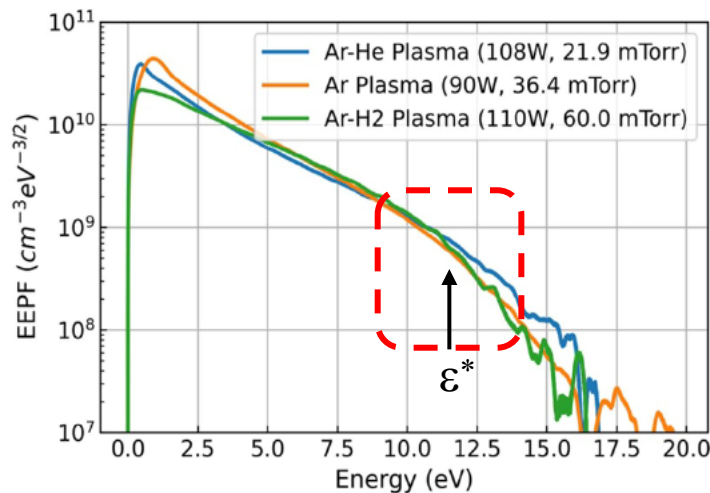
EEDF measured in argon CCP in a wide range of argon pressure at 13.56 MHz

Probe diagnostics based on the ion part of probe characteristic

(Single, double and triple probes)



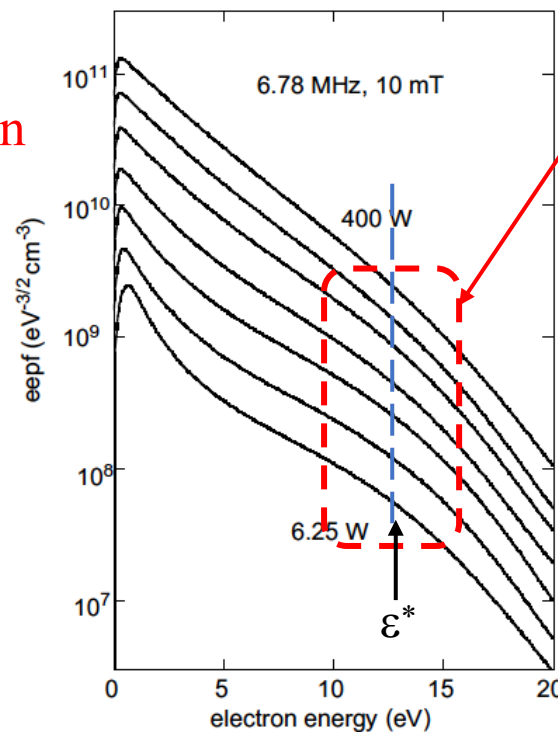
Uncertainty in electron temperature is due to $V_{pfl} \approx \epsilon^*/e$ where EEPE changes its slope $\propto T_e^{-1}$, arbitrariness in the ion current approximation $I_i(V_p)$ and unknown plasma potential.



The electron temperature T_e is found the I/V slope

$$T_{ei} = eI_e[dI_e/dV]^{-1} = e(I_p - I_i)[dI_p/dV - dI_i/dV]^{-1}.$$

Plasma density n is found using I_p , V_p , V_{fl} , T_e from ion current orbital motion theory (OMT for $a_p < \lambda_D$), and radial motion theory (RMT for $a_p > \lambda_D$), or from their modifications, like Laframboise theory.



Area of the floating potential

It is unknown whether the floating potential V_{pfl} is larger or smaller than ϵ^*/e and which T_e is measured.

There is no such problem with a Maxwellian EEPF where the slope is constant.

Comparison n/n_0 obtained with different method. n_0 - from EEDF

Plasma density equality factor n/n_0 . n_0 was found as appropriate integral of the measured EEDF

Source	Gas/pressure	EPPC	Ion OMT	Ion RMT	Cut-off pr.	Hairpin pr.
Ref. 10	Ar, 30 mTorr	1.34	2.5	0.16		
Ref. 10	Ar, 0.3 Torr	0.38/0.07	3.3	1.4	~1.4	
Ref. 18	He, 40 mTorr	0.85	9	0.25		
Ref. 21	Ar, 7–22 mTorr		2.6–3.25		1.1	1.5
Ref. 19	Ar, 1 mTorr		≈ 3	0.3		
Ref. 19	Ar, 10 mTorr		≈ 4	0.45		
Ref. 19	Ar, 0.1 Torr		≈ 2	0.14		

The reason for huge error in the plasma density obtained from ion current to the probe is due to unrealistic assumptions in the ion current theories $I_i(V_p)$.

The error in electron temperature is due to $V_{pfl} \approx \varepsilon^*/e$ where EEPE changes its slope $\propto T_e^{-1}$.

1. Systematic analysis of plasma density measurements carried by different author groups showed that n calculated from OMT are few times larger than n_0 , while those found using RMT are few times smaller.
2. In the case when EEDF is close to Maxwellian one at $\varepsilon < e^*$, n calculated from EPC is close to that found from cut-off, hairpin probes and EEDF.
3. The best agreement in evaluation of n was found between Cut-off and EEDF methods having less (than others methods) assumptions.

Why Probe diagnostics based on ion current is so inaccurate?

Ion current theory assumptions:

1. Maxwellian EEDF & IEDF, or $T_i = 0$.
2. No ion collisions in disturbed by the probe area within orbital zone ($R_0 \ll \lambda_i$), or probe sheath ($R_{sh} \ll \lambda_i$).
3. 1-D geometry of the disturbed zone.
4. No ion drift in ambipolar field.
5. Knowledge of the plasma potential.
6. Knowledge of the ion temperature.

Some wide spread myths about ion current diagnostics:

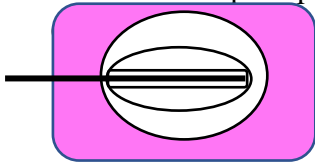
1. $n(I_p, V_p)$ is the ion density n_i
2. Ion part of probe I/V is immune to RF

RMT should work for a very dense plasma at mTorr pressure range ($\lambda_i \gg a_p > \lambda_D$), and Maxwellian EEDF up to $\varepsilon > eV_{fl}$.

These assumptions are usually not applicable in real plasma experiments.

1. No Maxwellian EEDF & IEDF. The ion current is defined by T_{el} , while measured T_e corresponds to T_{he} ; $T_{el} \neq T_{he}$. $f(\varepsilon)!!!!$
2. RMT & OMT are extremely sensitive to ion collisions limiting maximal gas pressure to few mTorr. **note**, $\lambda_i \ll \lambda_e$.

3. At high probe voltage, the sheath & orbital zone became spherical having different $I_p(V_p)$ scaling than cylindrical

4.  $I_p \propto V_p^{1/2}$ for cylindrical
 $I_p \propto V_p$ for spherical

4. In gas discharge, $v_{idrift} \gg v_{thermal}$.

5. $V_{fl} \neq (T_e/2e)\ln(M/2\pi m)$ cyl. probe and non-Maxwellian EEDF, $T_{el} \neq T_{eh}$. V_{fl} is extremely sensitive to shape of $f(\varepsilon)$.

Concluding remarks on Langmuir probe diagnostics

Electron part of probe current

- Non Maxwellian EEDF
- Uncertainty in plasma potential
- Arbitrariness in ion currents approximation affect T_e of fast electrons
- Gives accurate plasma parameters when EEDF close to Maxwellian in the elastic energy, $e < e^*$
- Assumption of collisionless regime (assumed in the theory) is valid for gas pressure 2 orders of magnitude higher than that for Ion current diagnostics, since $\lambda_e \gg \lambda_i$.
- There are few efforts to build probe theory accounting for magnetic field and collisions



Ion part of probe current

- Very large inaccuracy in T_e and n for the reasons considered above
- Used in practice theoretical RMT, OMT and Laframboise theory are not applicable at gas pressure higher than few mTorr
- At low gas pressure, high plasma density, when $l_e \gg \lambda_D < r_p$, and EEDF is Maxwellian for electron energy $\varepsilon \leq \varepsilon^* + [\varepsilon]$, The RMT may give the correct number for T_e and n
- There are theories to account for ion collisions in the ion probe theory, but they were used in experiment only by the authors



Unfortunately, these efforts have very little support

EEDF measurement in gas discharge plasmas

Since EEDF in gas discharge plasma, as a rule, is non-Maxwellian, the EEDF measurement according to Druyvestein method is the only reliable probe diagnostics.

According to Druyvestein, $\frac{d^2 I_e}{dV^2} = \frac{e^2 S p}{4} \sqrt{\frac{2e}{mV}} F_e(\epsilon) = \frac{e^2 S p}{2\sqrt{2}} f(\epsilon)$, where $f(\epsilon) = \epsilon^{-1/2} F_e(\epsilon)$ is the electron energy probability func.

$$n = \int_0^\infty \sqrt{\epsilon} f(\epsilon) d\epsilon \text{ and } T_{\text{eff}} = T_e = \frac{2}{3} n^{-1} \int_0^\infty \epsilon^{\frac{3}{2}} f(\epsilon) d\epsilon. \quad \text{Similarly, for } z, v^*, v_{en}, v_{ee}, v_{ei}, \lambda_D, v_s \text{ and so on.}$$

$F_e(\epsilon)$ is EEDF, $f(\epsilon)$ is EEPF that for Maxwellian distribution looks as a strait line.

$$\text{Analog AC: } I_e(V_0 + \delta) = I_e(V_0) + \delta \frac{dI_e}{dV} + \frac{1}{2} \delta^2 \frac{d^2 I_e}{dV^2} + \dots \quad \delta = \delta(t) \ll T_e$$

$$\delta = \delta_0 \sin \omega t \quad \text{Second harmonic, } 2\omega$$

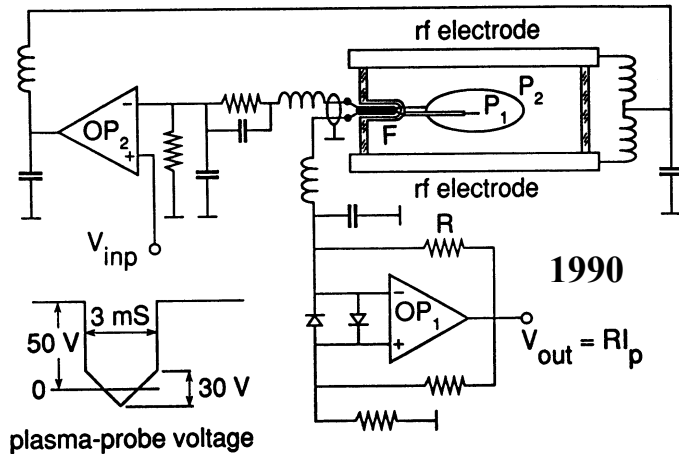
$$\delta = \frac{1}{2} \delta_0 [1 + \cos \omega t] \sin \Omega t \quad \text{Envelop frequency, } \omega$$

$$\delta = \frac{1}{2} \delta_0 [\sin \Omega t + \sin \omega t] \quad \text{Beating frequency, } \Omega \pm \omega$$

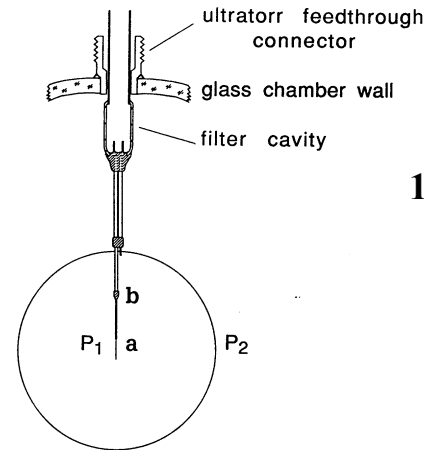
$$\frac{d^2 I_e}{dV^2} \propto \delta^2 \ll (T_e/e)^2$$

Large δ results in higher sensitivity, but worse energy resolution

Acquisition and processing of probe characteristic

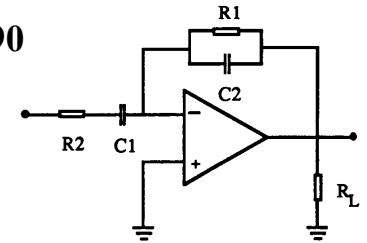


1990



Differentiation circuit with filtering (smoothing)

1990

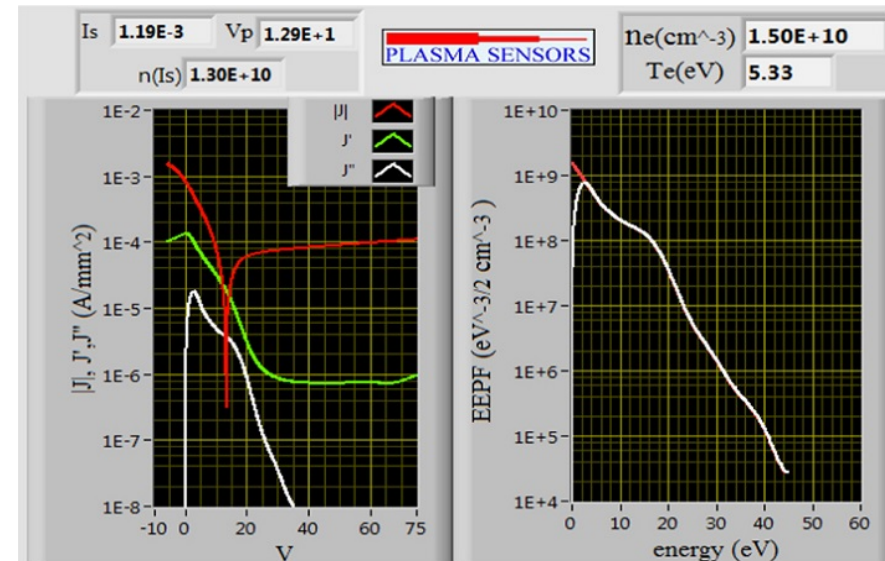


Compensation for rf (ω , & 2ω), lf (<100 kHz) and dc (< 400 V) plasma potential).

Measurement, processing and display takes (1-10) Sec.



Comparison of 4 commercial probe systems see in JAP, **118**, 233302 (2015)

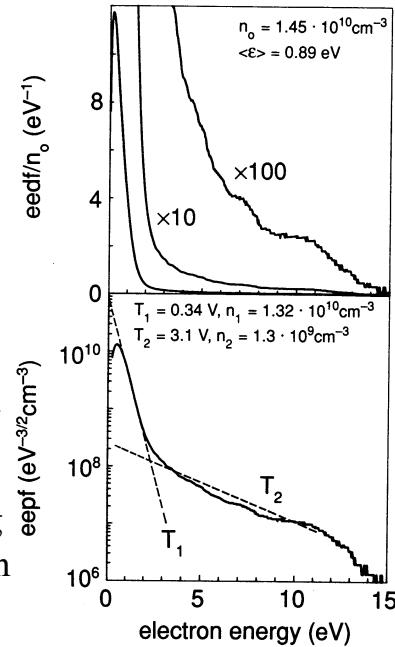


EEDF measured in Ar & He CCP

EEDF measurements in CCP with high energy resolution ($\Delta\epsilon < T_e/2$) and high dynamic range ($f_{\max}/f_{\min} > 10^3\text{-}10^4$) revealed some few electron kinetic phenomena in rf discharges

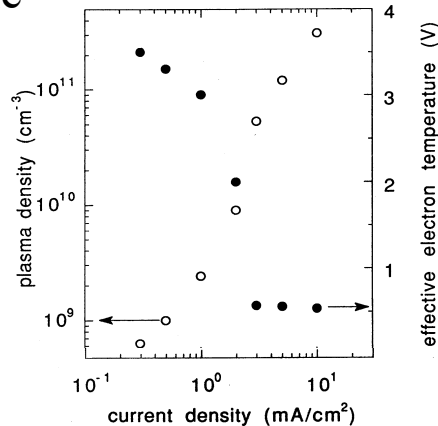
1. CCP heating mode transition

Ar

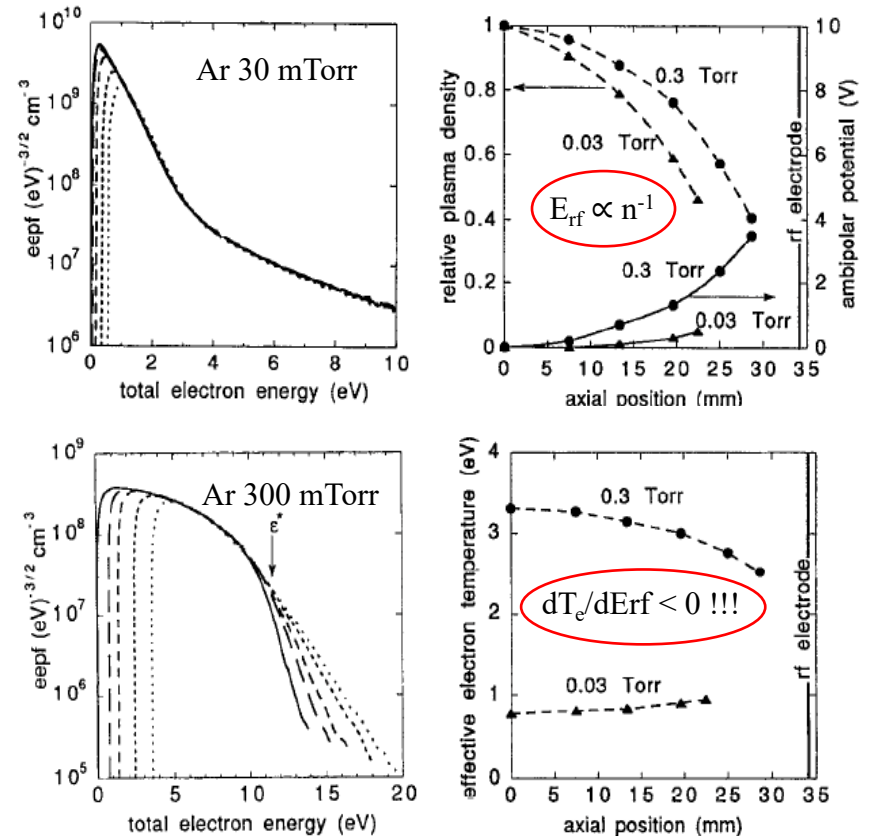


2. CCP transition to the γ -mode

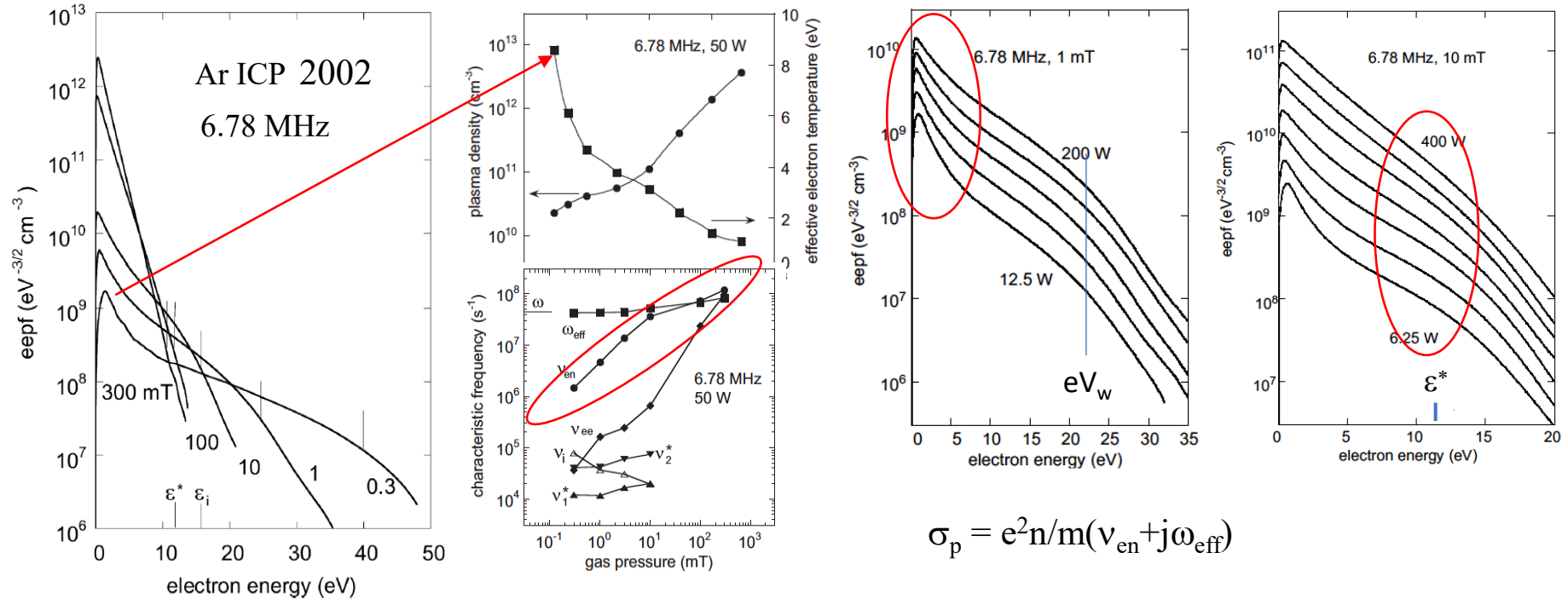
He



3. Paradoxical $T_e(x)$ distribution in Ar CCP

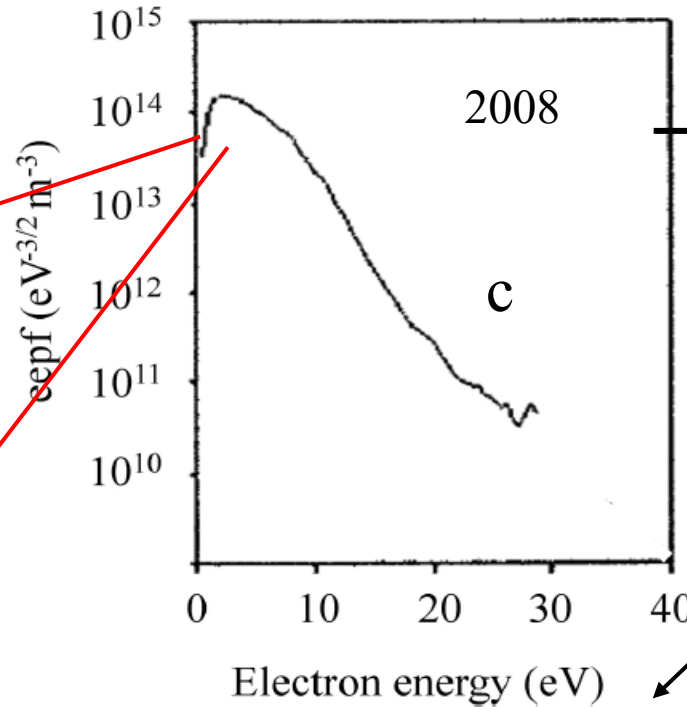
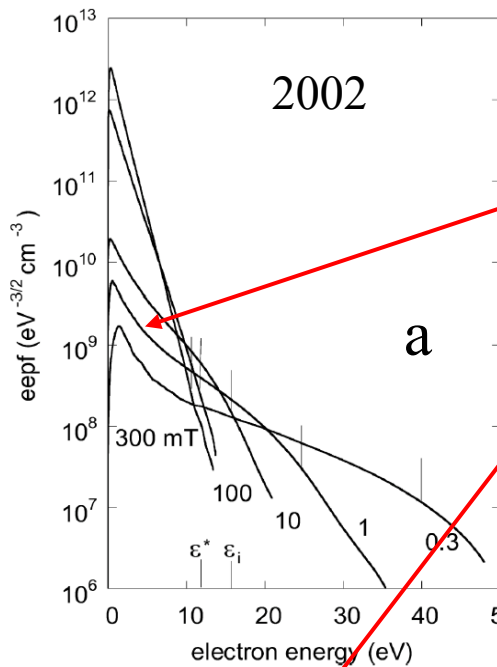


EEPF measured in Ar ICP



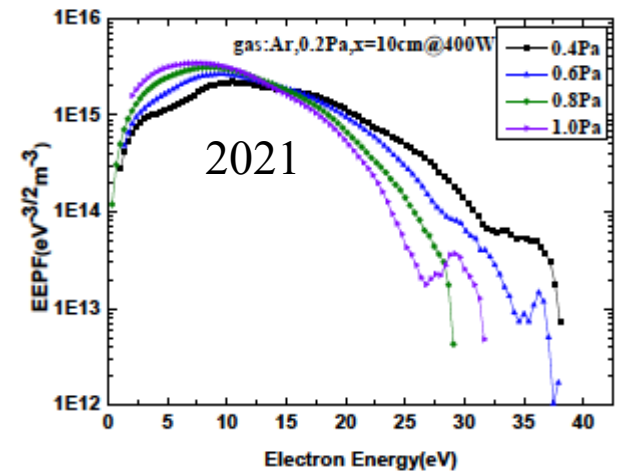
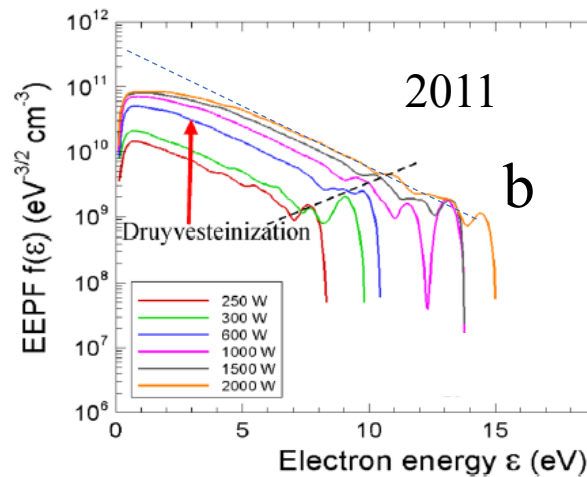
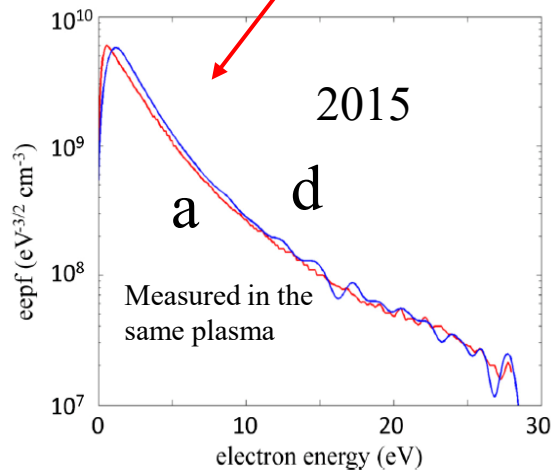
- EEPF measured with $\Delta\epsilon \ll T_e/2$ and dynamic range over $10^3 - 10^4$
- EEPF is concave at low energy ($\epsilon < \epsilon^*$) and low plasma density n . With growing n , the EEPF Maxwellize ($\nu_{\text{ee}} \propto n\epsilon^{-3/2}$). This shape of EEPF was later revealed in numerous numerical simulation, but the majority of published experiments in ICP shows very different EEPF shape
- Electron-neutral collision frequency $\nu_{\text{en}} \propto p$ at $p \leq 10$ mTorr and $\nu_{\text{en}} \propto p^{1/2}$ at $p \geq 10$ mTorr due to Ramsauer effect and T_e drop with gas pressure

“Progress” in EEDF measurement in Ar ICP at low gas pressure



“...is by far the best commercial Langmuir Probe on the market, ...”

The majority of electrons are missing in these measurements



Main problems in EEDF measurement

Due to error augmentation inherent to differentiation procedure, even small error in $I_p(V)$ measurement can result in enormous distortion in the inferred $f(\varepsilon) \propto I_p''(V)$

Distortions in the probe characteristic $I_p(V)$ are caused by:

- Probe contamination (resulting in stray resistance R_c). Needs probe cleaning
- Change of probe work function (up 1.5 V). Needs fast ($< \text{few ms}$) probe scan
- Low and high frequency noise (probe non linearity). Needs dc, lf and rf compensation
- Probe circuit resistance due to wall coating and sheath, rf filters and so on
- Violation of basic requirements and limitations of the probe methods (too large gas pressure, excessive probe size and/or its holder, ...)

These distortions are usually not visible in probe characteristic, but well seen in the resultant $f(\varepsilon)$ that allows, at glance, to see the problem. Therefore, for reliable $f(\varepsilon)$ measurement, a significant attention has to be paid to accuracy in $I_p(V)$ measurement and to ability to monitor $f(\varepsilon)$ in the real time.

The main problem in the majority of publications is a large gap $\Delta\varepsilon$ between zero and maximum of the measured $f(\varepsilon)$, $\Delta\varepsilon > T_e/2$, thus, missing information about the majority of electron population (Effect of EEDF Druyvesteinization).

Machine learning

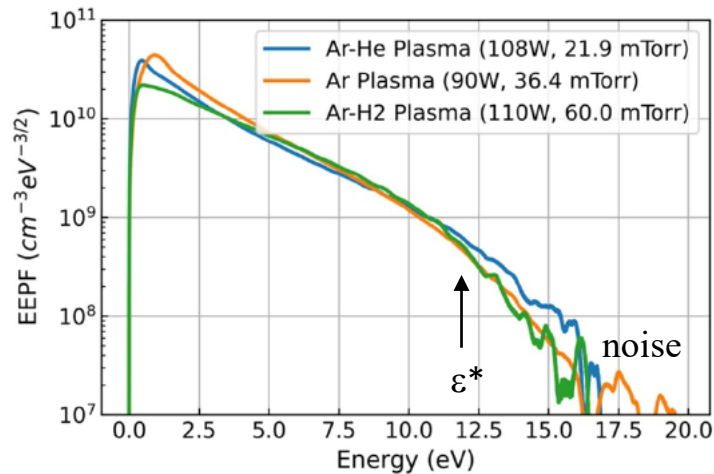


Figure 2. Example of measured electron energy probability functions (EEPFs) for Ar, Ar-He and Ar-H₂ mixtures, at various pressures and input powers.

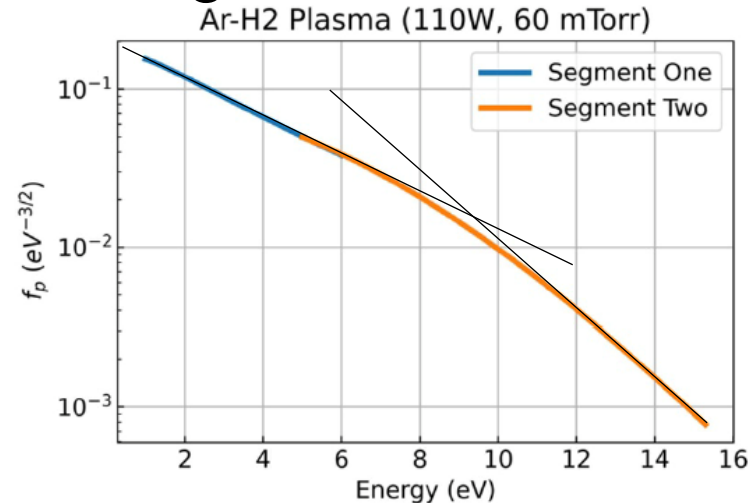


Figure 3. Example of a post-processed and fitted electron energy probability function f_p .

- Low energy peak and slope change at $\epsilon = \epsilon^*$ reflect the specific of electron stochastic heating and effect of EEDF slope change due to inelastic electron collision (excitation)
- At increased rf power and gas pressure, Druyvesteinization occurs
- Due to threshold-like onset of excitation at $\epsilon > \epsilon^*$, the experiment shows the change in $\ln f(\epsilon)$ slope at $\epsilon \approx \epsilon^* = 11.56$ eV, but the “educated” computer shows a very smooth transition around $\epsilon \approx 9$ eV.
- Such smoothing can be done for few second by hand, or instantly by elec-tronic or digital filtering, instead of teaching computer to own ignorance.

Conclusion Remarks

- The main problems in probe diagnostics are: neglecting of basic requirements and limitations for probe for diagnostics, ignorance in gas discharge physics, and lack of engineering skill in design of home probe experiment and commercial instruments.
- There are also some fundamental limitations of classical probe methods associated with applicability of the probe theory to a real plasma, like non-Maxwellian $f(\epsilon)$ and particle collisions). Found with probe the plasma parameters may be in significant error, especially those found from the ion current to probe I/V using OM and RM theories.
- The measurement of EEDF (when it done professionally) is the most accurate probe measurement technique. But the majority of published EEDF are heavily distorted missing of low energy electrons that are the main portion of electron population
- These problems were addressed in many well cited, but not read reviews, thus, having little impact on quality of probe measurements in recent publications

Recommended
literature

J. Appl. Phys., **129**, 041101 (2021),

J. Appl. Phys. **118**, 233302 (2015),

Phys. D: Appl. Phys. **44**, 233001 (2011)

Thank you