Shining a Light on the Complex Physics of Low-Temperature Plasmas

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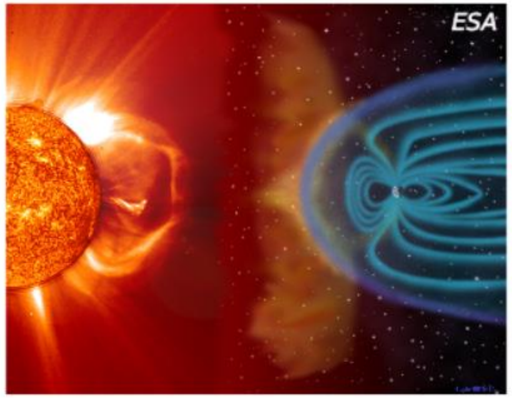
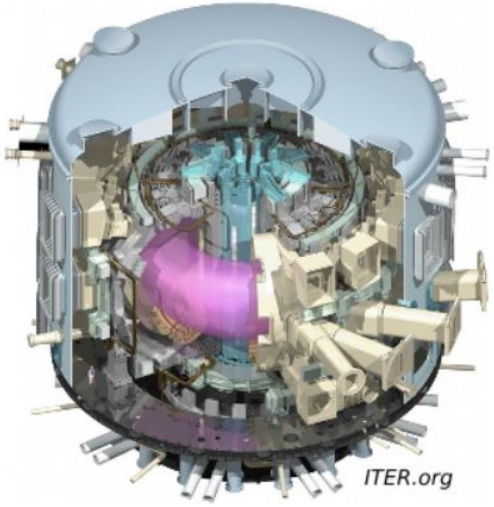
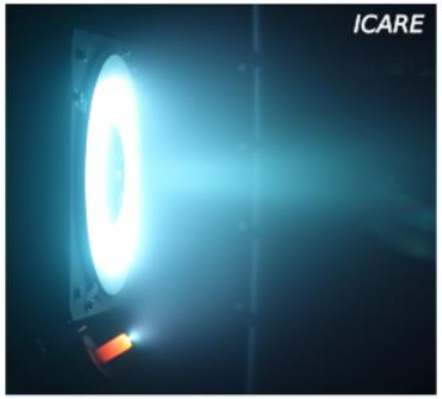
Introduction

- several exciting areas have emerged in LTPs in recent decades:
  - microplasmas, nanostructuring and materials fabrication, metamaterials and photonic crystals, medical therapies...

- many long-standing questions relevant a range of plasmas can now be addressed using new tools: experimental and numerical

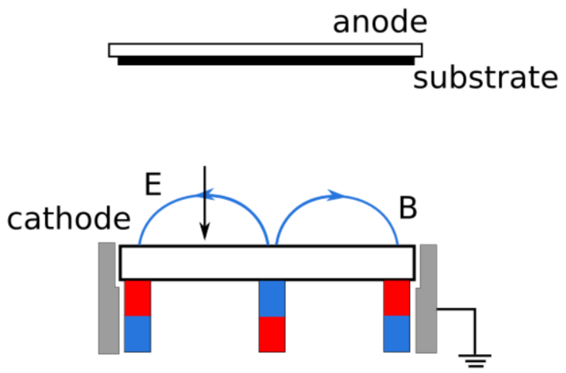


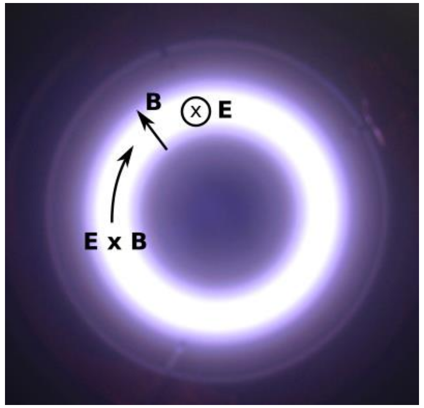




Progress in plasma-assisted deposition

- emergence of the now well-known HiPIMS (high power impulse magnetron sputtering) regime for **planar magnetrons**
  e.g. Kouznetsov et al, Surf. Coat. Technol. 122, 290 (1999)





planar magnetron ExB geometry

significance

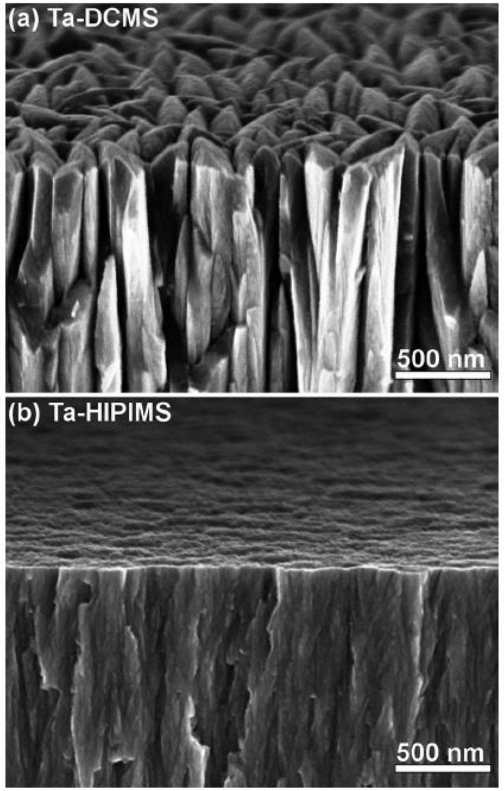
access to improved thin film characteristics: density, grain orientation, adhesion...

challenges

dense plasma ($10^{18} – 10^{20}$ / m^3), transient regime, 3D physics

⬇

difficult to model



Fager et al, J. Appl. Phys. 121, 171902 (2017)

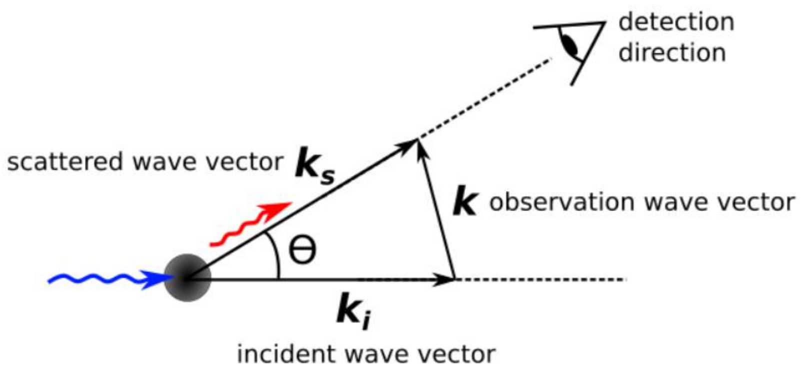
Progress in plasma-assisted deposition

- the promise offered by this device is an incentive to advance modeling and experimental tools

- ideally, we wish to exploit methods which are:
  - non-invasive, non-perturbative
  - spatially-, temporally- resolved

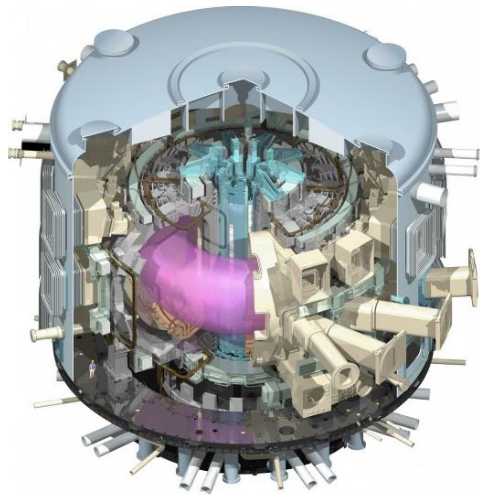
  ➡ optical diagnostics

- Thomson scattering: elastic scattering on free charged particles



$$\mathbf{k} = \mathbf{k}_s - \mathbf{k}_i$$
$$\omega = \omega_s - \omega_i$$

$$|k| \cong 2\,|k_i|\,\sin(\theta/2)$$



a vital tool for fusion studies

Information from Thomson scattering

$\ell < \lambda_D$: incoherent regime

- individual random (thermal) fluctuations visible

$$\frac{d^2P}{d\Omega_s d\nu_s} = \left[2\pi r_e^2 \int_V \langle S_i \rangle \, d^3r \, |\hat{\mathbf{s}} \wedge (\hat{\mathbf{s}} \wedge \hat{\mathbf{e}})|^2\right] f_k\left(\frac{\omega}{k}\right)\frac{1}{k}$$

$f_k(v_k) =$ 1D velocity distribution function along $\mathbf{k}$

Maxwell-Boltzmann velocity distribution

$$f_k = \underline{n_e}\left(\frac{m_e}{\underline{2\pi T_e}}\right)^{1/2} \exp\left(-\frac{m_e {v_k}^2}{\underline{2T_e}}\right)$$

$\ell >> \lambda_D$: coherent regime

- collective, non-random fluctuations visible

$$\frac{d^2P}{d\Omega_s d\nu_s} = \frac{r_e^2 P_i}{A} |\sqcap \cdot \hat{\mathbf{e}}|^2 n_e V S(\mathbf{k}, \omega)$$

$S(\mathbf{k}, \omega) =$ scattering form factor

$$S(\mathbf{k}, \omega) = \frac{1}{n_e T V}|N_e(\mathbf{k}, \omega)|^2$$

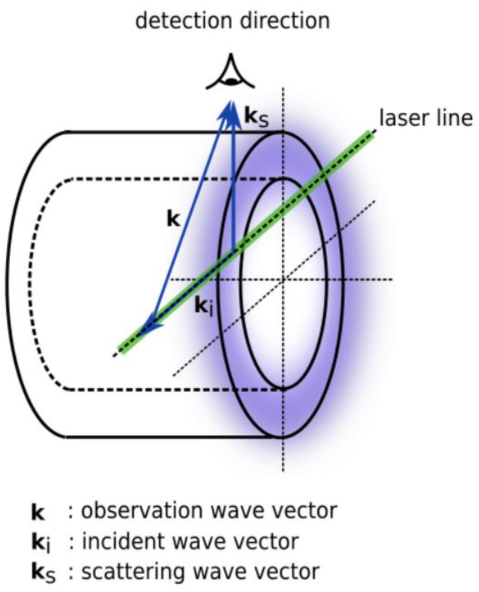
$N_e(\mathbf{k}, \omega) =$ Fourier transform of electron density

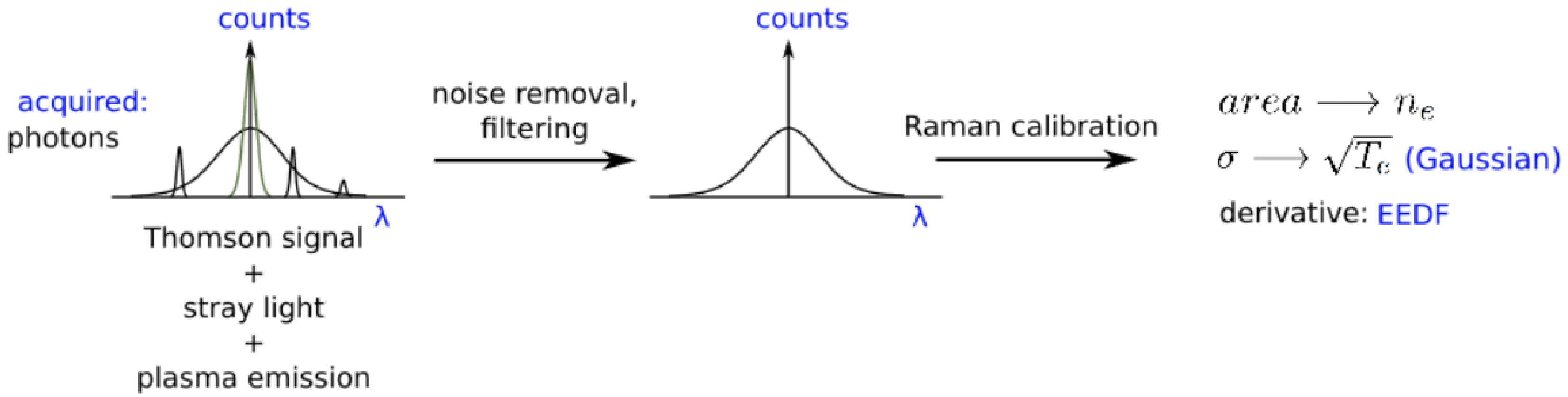
$\Omega_s =$ solid angle of detection
$V =$ scattering volume
$\hat{\mathbf{s}}, \hat{\mathbf{e}} =$ unit vectors in scattering and incident field directions
$\langle S_i \rangle = P_i / A =$ mean incident Poynting vector

$\sqcap =$ tensor polarization operator

Electron property measurement with ITS

- Incoherent Thomson scattering (ITS)
  - challenges:
    - low density, few photons
    - stray light and other signals



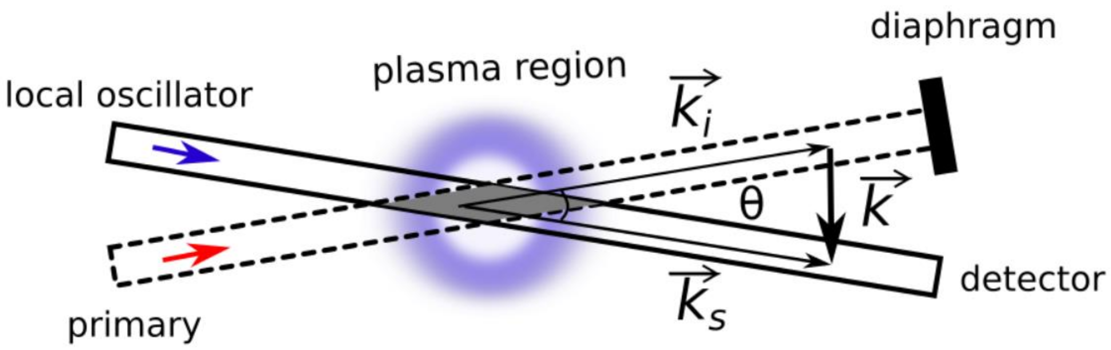


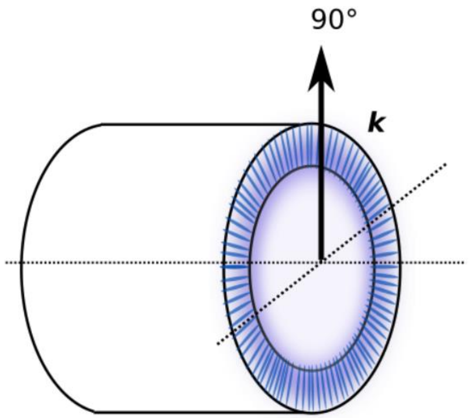
- over the past few years, we've been able to lower the detection threshold to 10^{16} /m^3

  ➡ new range of diagnostic studies accessible

Fluctuation measurement with CTS

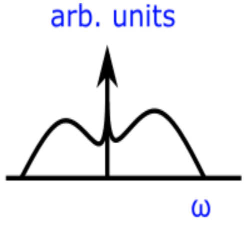
- Coherent Thomson scattering (CTS)
  - challenges:
    low density, more complex setup



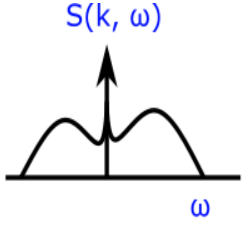


- signal analysis (real amplitude, frequency content)

acquired:
time-varying signal
complex signal → FFT of complex signal; noise removal →



scattered signal x *M* →

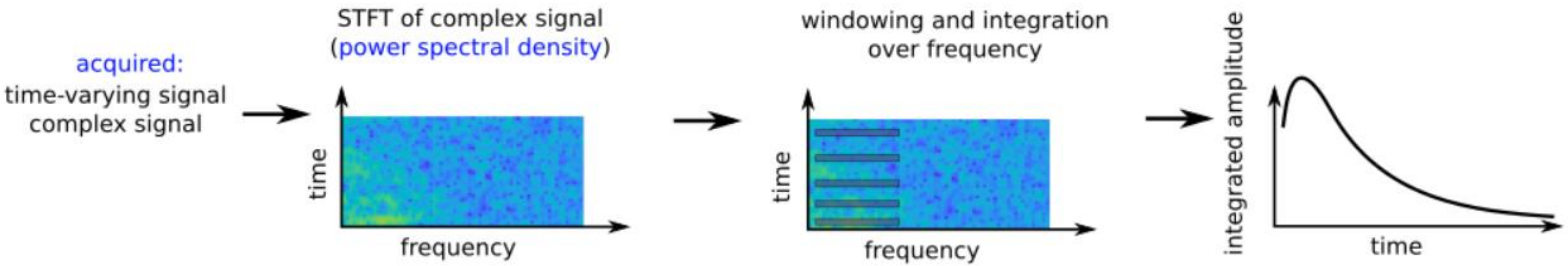


dynamic form factor

integration → static form factor S(k)

Fluctuation measurement with CTS

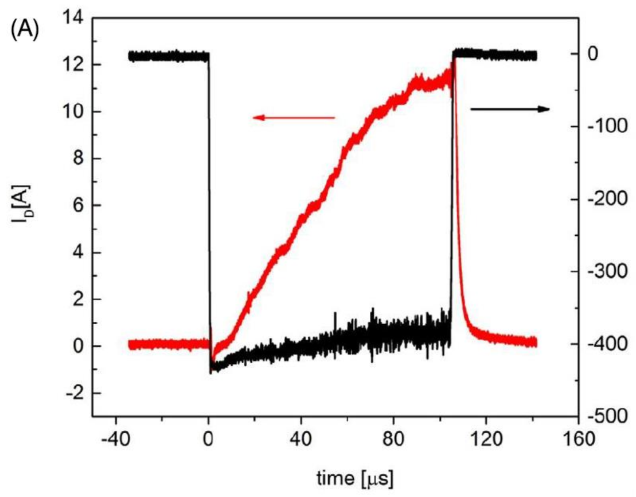
- signal analysis (preservation of temporal features)



- sophisticated studies of plasma turbulence at scales inaccessible to other diagnostics are possible

HiPIMS plasma features

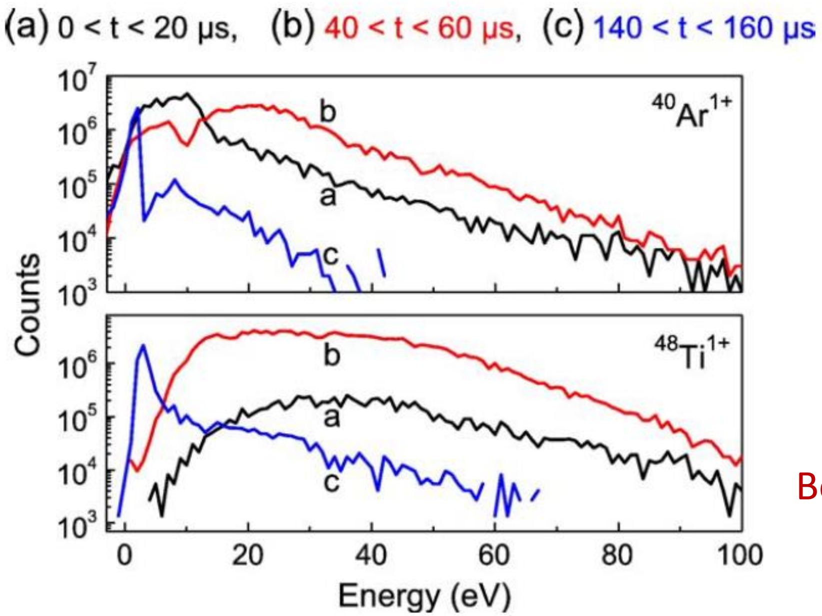
- planar magnetron operation: rapidly-varying current profiles



High Power Impulse Magnetron Sputtering: Fundamentals, Technologies, Challenges and Applications
ed. Lundin, Minea and Gudmundsson, Elsevier (2020)

- rapidly-evolving species content and dynamics

  ion energies



Bohlmark et al, Thin Solid Films 1522, 515 (2006)

Progress in plasma-assisted deposition: modeling

- in recent years, a number of codes have emerged to meet the needs of the HiPIMS regime

- self-consistent PIC-MCC in 2D for short pulses

Revel et al, PSST 27, 105009 (2018)

- PIC-MCC in 2D and 3D (linear magnetron)

Pflug et al, Surf. Coat. Tech. 260, 411 (2014)

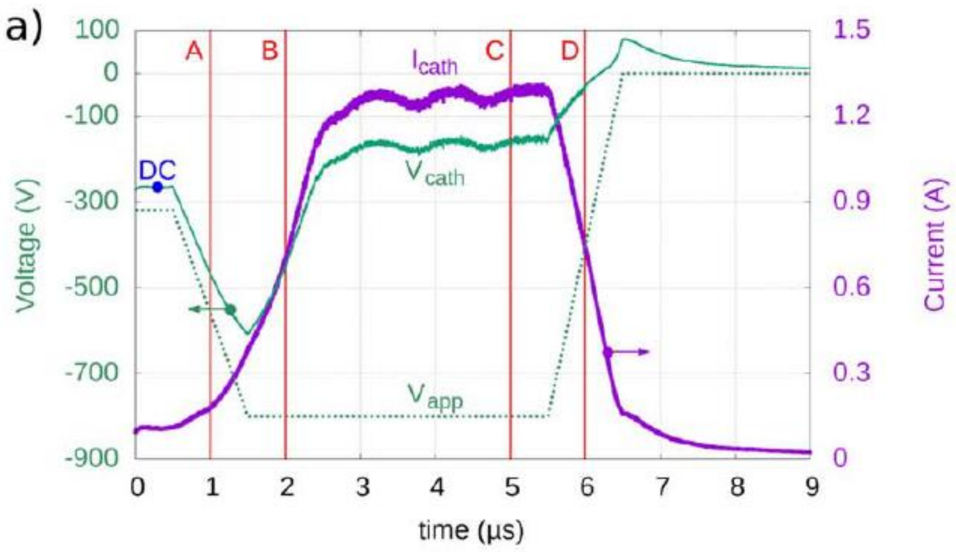
- ionization region global model

Raadu et al, PSST 20, 065007 (2011)

- bulk plasma model

Brenning et al, PSST 17, 045009 (2008)

9 µs discharge pulse: 3 weeks parallel computing on 60 cores, each operating at 2.1 GHz



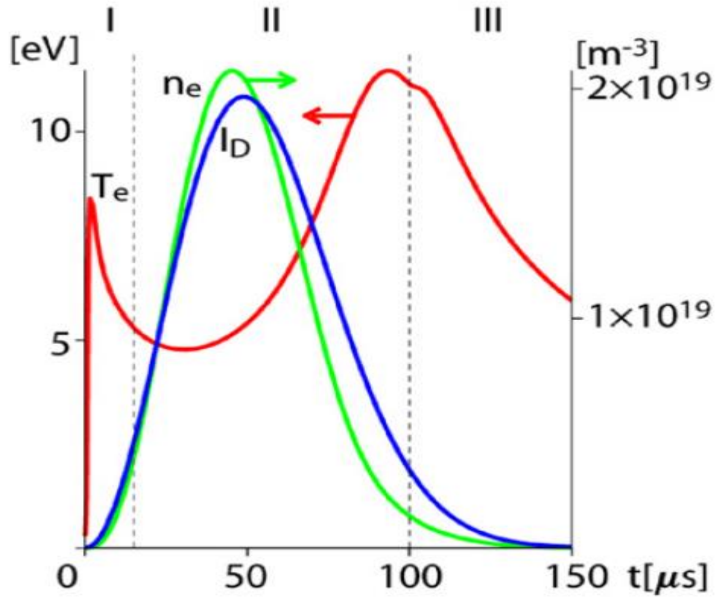
simulated current and voltage profiles
Revel et al.

- none yet has the capability to fully capture all the multiscale (temporal and spatial) physics of the HiPIMS regime

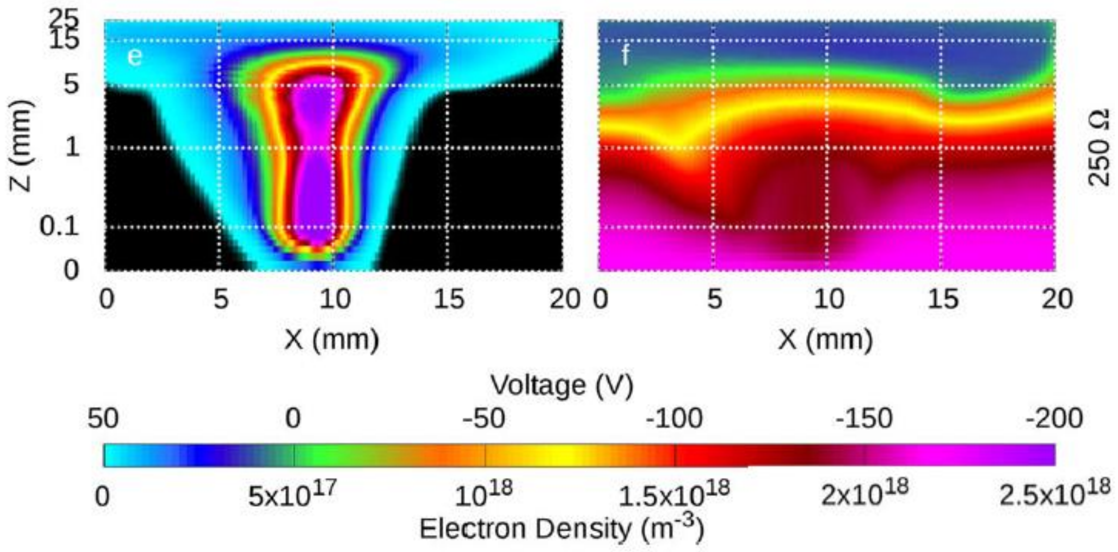
Progress in plasma-assisted deposition: modeling

- electron property determination in codes used in HiPIMS

Raadu et al, PSST 20, 065007 (2011)



Revel et al, PSST 27, 105009 (2018)



- given that every existing code has limitations, worth validating their outputs with diagnostics

Langmuir probes: unsuited for near-cathode region

OES: widely used, but line integrated + emission models required

THz: density measurements

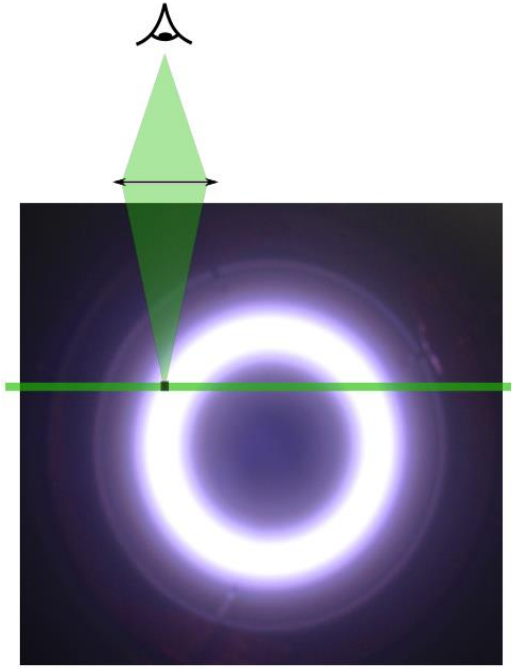
A Ando et al, J. Phys.: Conf. Ser. **227** 012016 (2010)

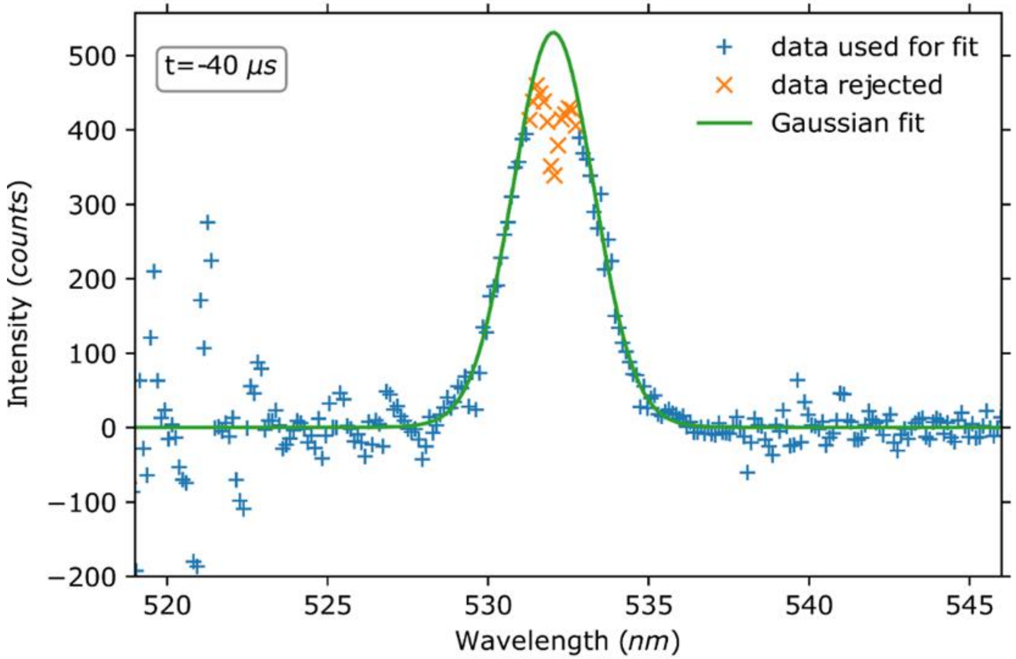
Meier et al, PSST 27 035006 (2018)

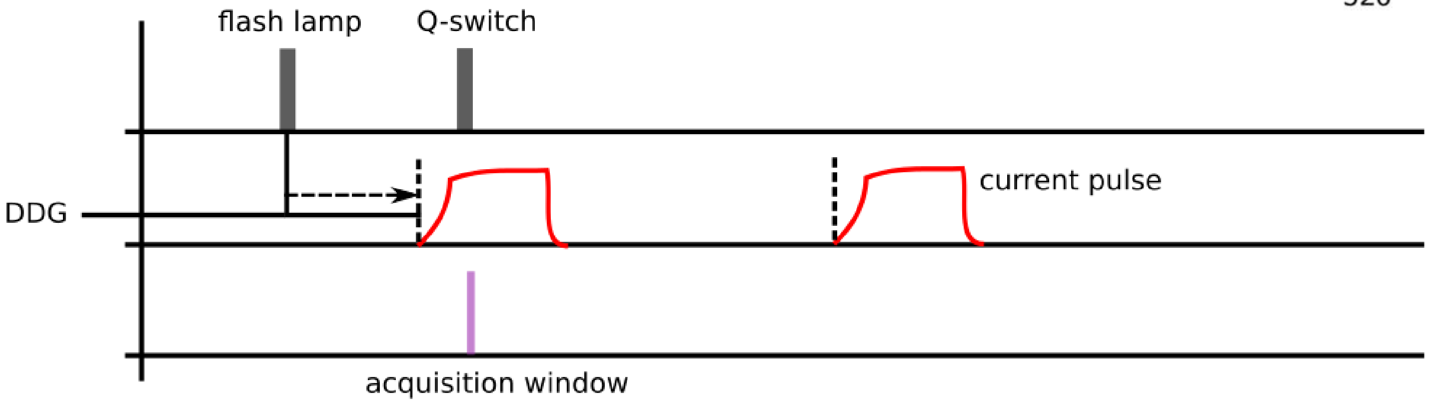
Progress in plasma-assisted deposition: ITS

- electron property determination with ITS:

  access to highly-resolved spatially, temporally-resolved information





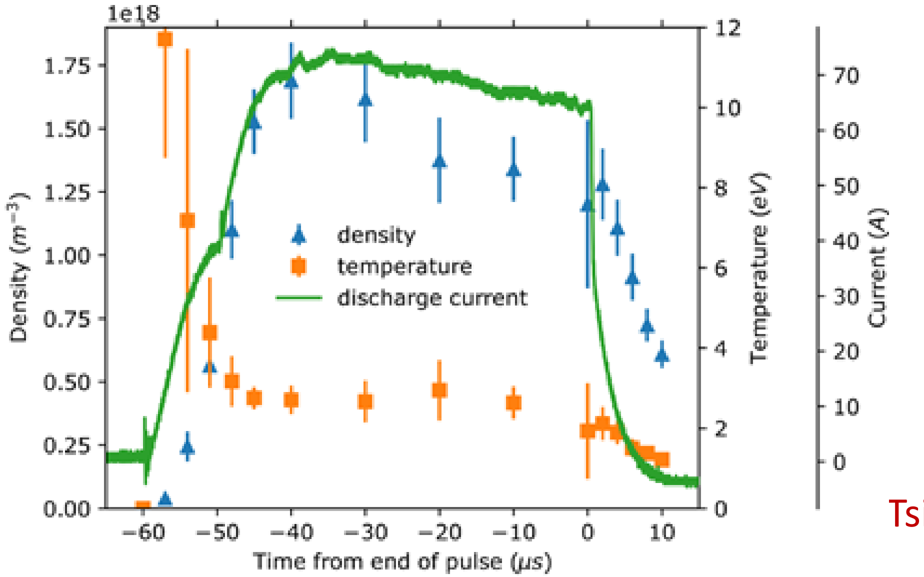


Tsikata et al, PSST 28, 03LT02 (2019)

Progress in plasma-assisted deposition: ITS

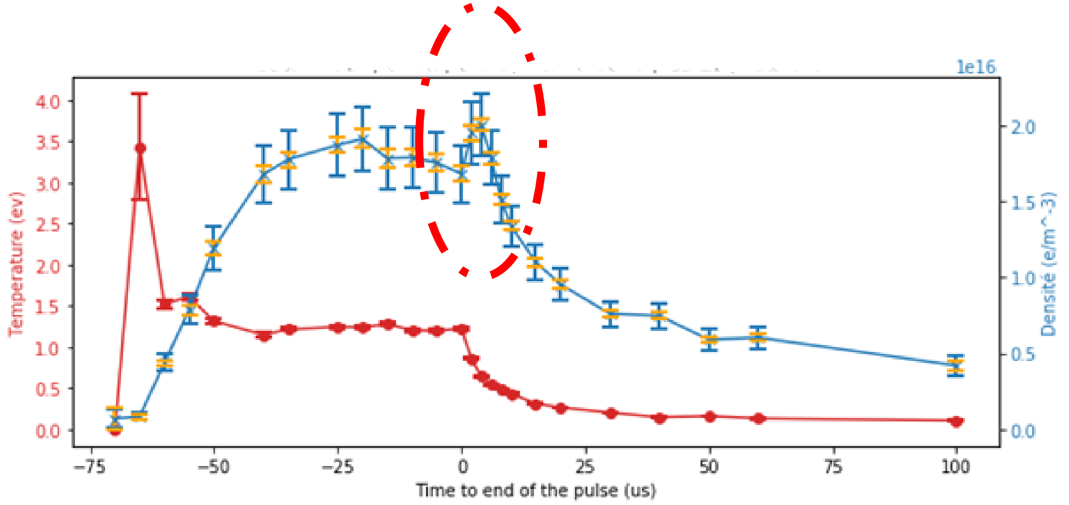
- electron property determination with ITS

Ar working gas,
Ti cathode



- map of electron properties from discharge initiation into post-discharge

Tsikata et al, PSST 28, 03LT02 (2019)



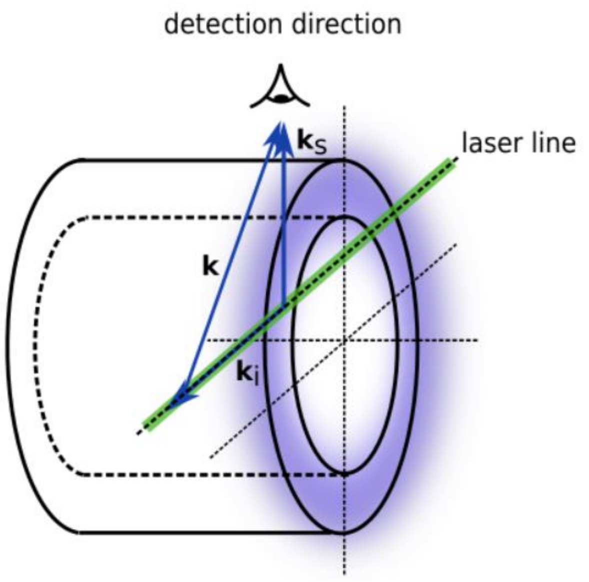
- evidence of Penning ionization at end of pulse

  $Ar^* + Ar \Rightarrow Ar + Ar^+ + e^-$

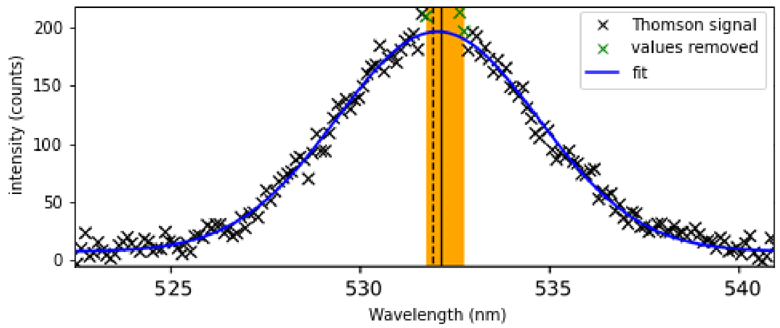
- access to this information can be used to validate recently-developed codes

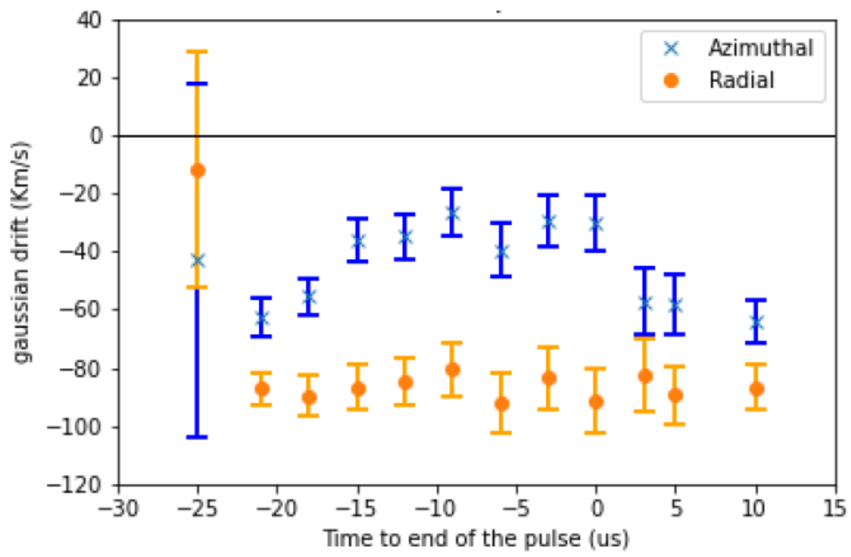
2020

Progress in plasma-assisted deposition: ITS



k : observation wave vector
$\mathbf{k_i}$: incident wave vector
$\mathbf{k_S}$: scattering wave vector





2020

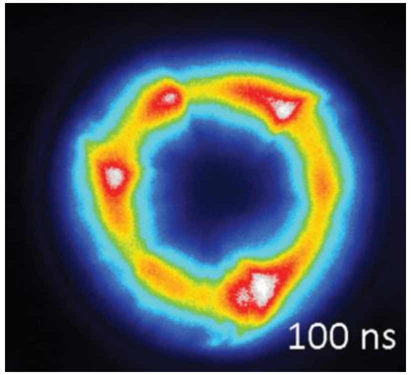
- expected ExB azimuthal drift, but significant radial drift directed towards center of cathode

- from these recent implementations, ITS can offer detailed information on local electron properties and dynamics (drift)

- other fundamental behavior of such magnetized discharges can be elucidated through CTS

Fluctuations in HiPIMS

- The vast majority of studies of fluctuations in planar magnetrons have focused on large-scale self-organization



diagnostics used:
- spectroscopic analysis
- probes

Anders et al, J. Appl. Phys. 111, 053304 (2012)

- azimuthal instabilities invoked to explain anomalously large discharge current

  observed: j_H/j_D = 2

  typical classical collision-based: j_H/j_D = 16 – 35

- measurement of fluctuations using probes, associated with modified two-stream instability (MTSI)

Lundin et al., PSST 17, 025007 (2008)

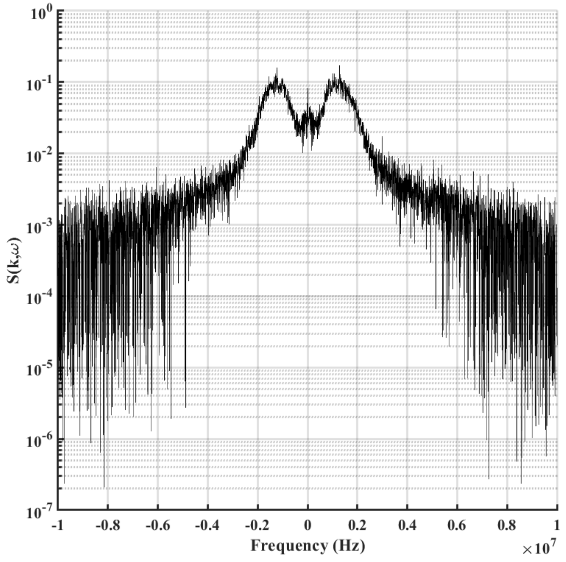
- recent studies linking spokes to anomalous transport

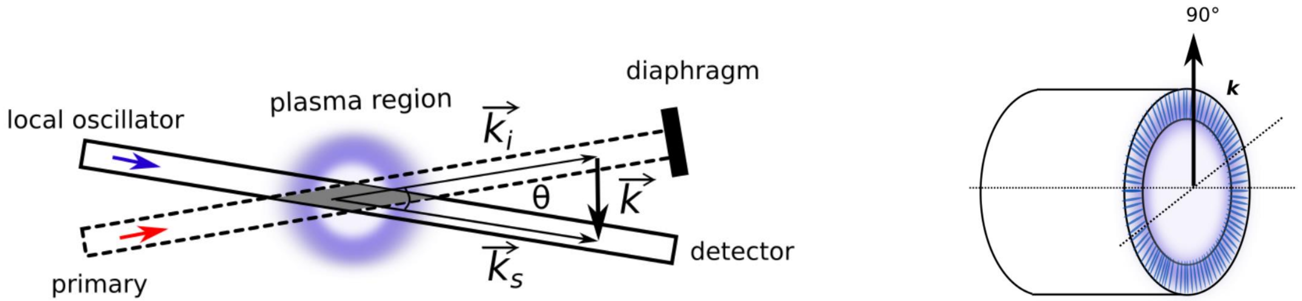
Hecimovic, J. Phys. D: Appl. Phys. 49, 18LT01 (2016)

Progress in plasma-assisted deposition: CTS

- measurements using CTS: presence of µscale electron density fluctuations compatible with the ECDI (electron cyclotron drift instability)

Tsikata and Minea, PRL 114, 185001 (2015)





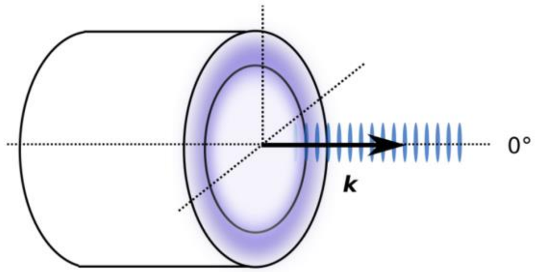
- observation wave vector aligned to capture electron density fluctuations in ExB drift direction
- simultaneous observation of both sides of the racetrack

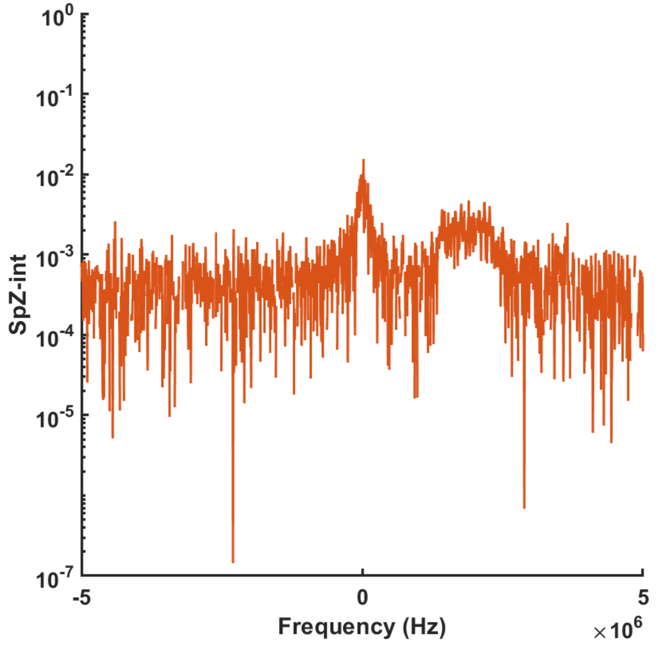
- note: unlikely to be a single instability responsible for transport
  - mode coupling (illustrated in simulations)

Studies of electron density fluctuations

- an axially-propagating wave also present: ion-ion two stream instability (IITSI)

Tsikata et al., Phys. Plasmas 21, 072116 (2014)
Hara and Tsikata, Phys. Rev. E 102, 023202 (2020)





- the IITSI is a special case of two stream instability: at least 3 species involved

dispersion relation

$$(k_\perp \lambda_D)^2 \left[1 - \frac{(1-\alpha)\omega_{pi}^2}{(\omega - \mathbf{k}\cdot\mathbf{U}_i^+)^2} - \frac{\alpha\omega_{pi}^2}{\left(\omega - \mathbf{k}\cdot\mathbf{U}_i^{2+}\right)^2}\right] + 1 - I_0(b)\exp(-b) + \sum_{n=1}^{\infty} \frac{2\omega^2 I_n(b)\exp(-b)}{(n\omega_B)^2 - \omega^2} = 0$$

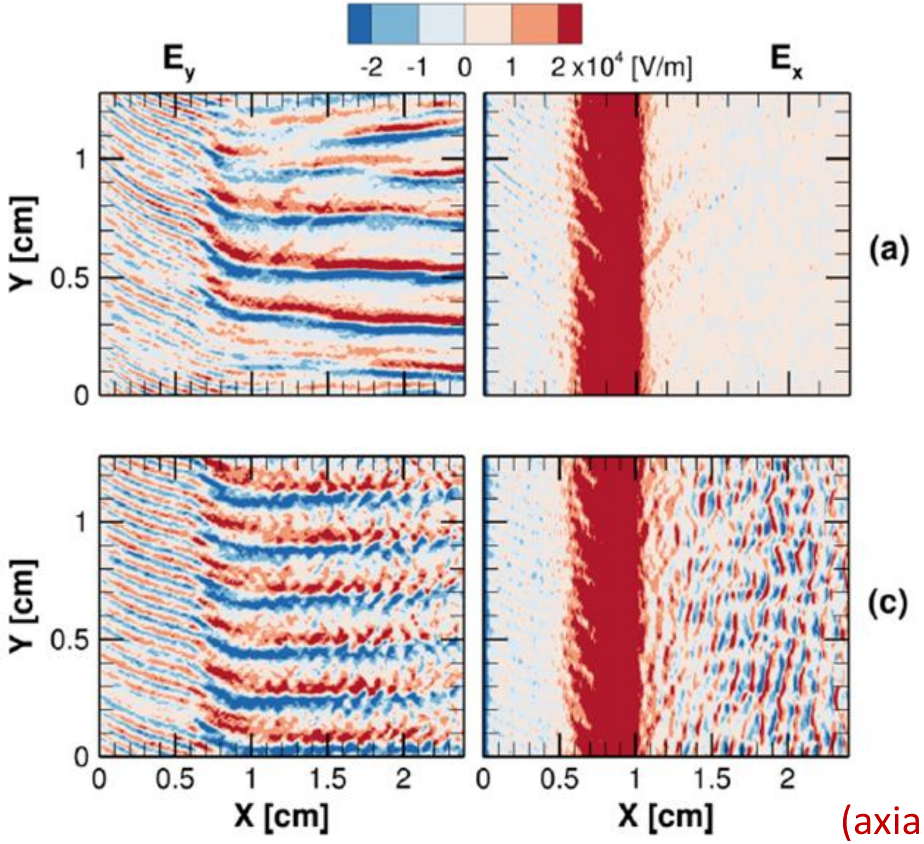
singly charged$^+$ | doubly charged^{2+} | magnetized electrons

Doubly charged ion fraction: $\alpha = \frac{2n_i^{2+}}{n_e}$

Studies of electron density fluctuations

- IITSI features first identified in thrusters: MHz frequencies, mm-length scales, primarily axial

azimuthal



axial

0 % Xe2+

20 % Xe2+
(axial modulation evident)

- coexistence with ECDI
- excited in the presence of Xe+ and Xe2+ populations

significance

- relevant to HiPIMS regime
  magnetized electrons, several ion species (gas and metallic)

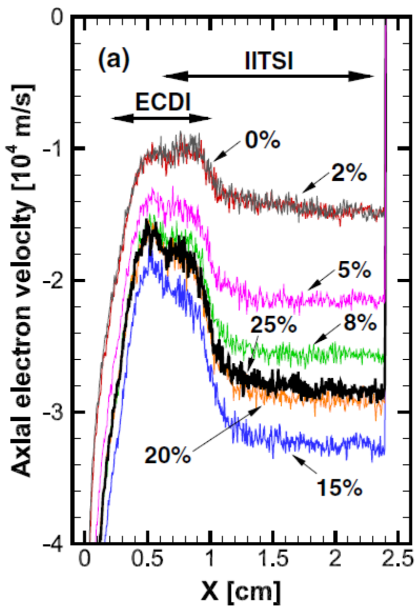
Hara and Tsikata, Phys. Rev. E 102, 023202 (2020)

Studies of electron density fluctuations

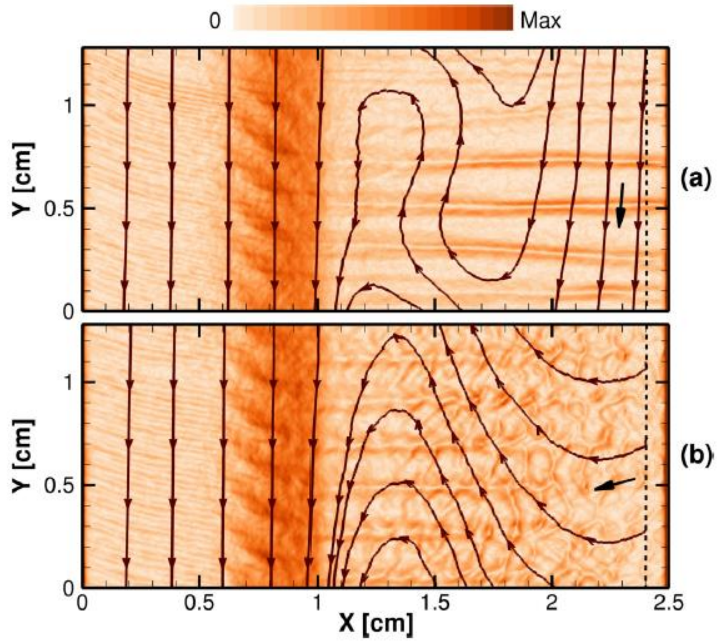
- electron transport was found to be increased by coupling of ECDI to IITSI:

  non-linear effects revealed by simulations

exit plane



azimuthal



ECDI alone

ECDI and IITSI

axial

transport increasing in presence of IITSI

electron streamlines directed more axially in presence of IITSI

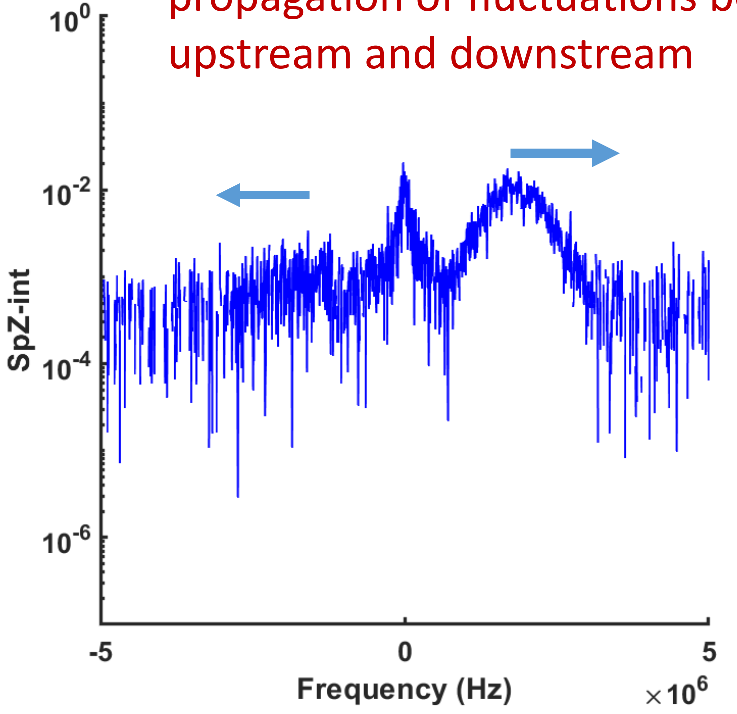
- electron transport in magnetrons will be determined by the contributions of several ion species

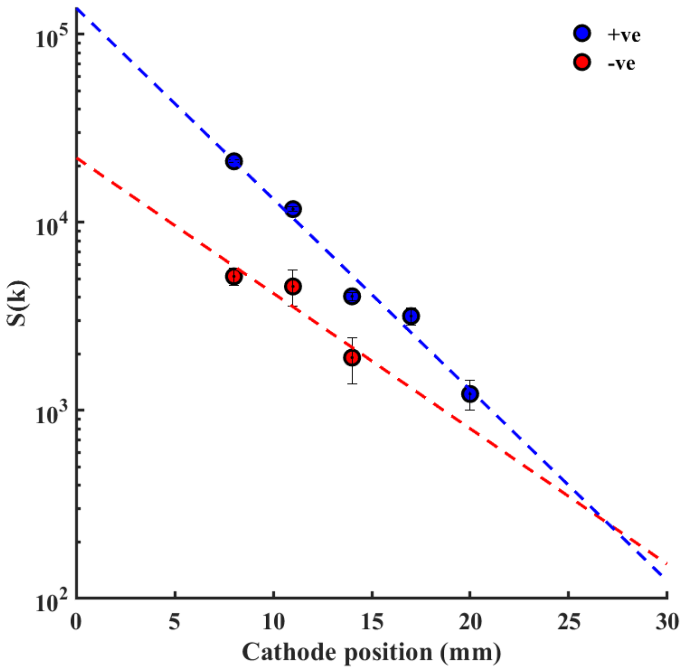
more challenging

Studies of electron density fluctuations

- in the HiPIMS regime of planar magnetrons, axial fluctuations (IITSI) have a distinct character

propagation of fluctuations both upstream and downstream





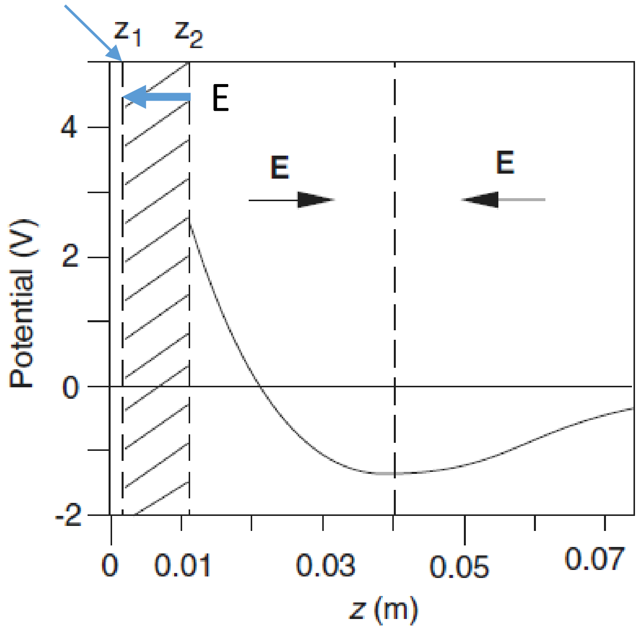
fluctuation amplitude rate of decrease differs

- do simulations show any related effects?

Studies of electron density fluctuations

- a feature unique to the HiPIMS regime: field reversal

ionization region



field reversal: pressure-driven current > discharge current

- in addition to several ion species, there are also varied directions of ion acceleration

➢ bulk plasma model

Brenning et al, PSST 17, 045009 (2008)

- experimental investigations into the propagation of fluctuations can offer insights into such phenomena

Conclusions

- advanced experimental implementations offer a wealth of insights into complex plasmas
  - particle properties
  - transport and plasma fluctuations

- in recent studies, we've sought to clarify physics of a particularly important technological cold plasma

- these studies provide a means to validate and improve physical models

as always, the physics looks more complex (and intriguing!) the more we examine it,
but our understanding is progressing rapidly 👍