White Paper for *Frontiers of Plasma Science Panel*

Date of Submission:	***18 June 2015***

	"P", "S"
• Plasma Atomic physics and the interface with chemistry and biology	
• Turbulence and transport	S
• Interactions of plasmas and waves	S
• Plasma self-organization	P
• Statistical mechanics of plasmas	

Indicate type of presentation desired at Town Hall Meeting:

	"X"
Oral	X
Poster	
Either Oral or Poster	
Will not attend	

Title:	**Self-organization in ExB discharges**
Corresponding Author:	Andre Anders
• Institution:	Lawrence Berkeley National Laboratory
• email:	aanders@lbl.gov
Co-Authors:	Edward Barnat (Sandia National Laboratories), Mark A. Cappelli (Stanford University), Jón Tómas Guðmundsson (Univ. Iceland), Michael Keidar (Georgetown University), Mark J. Kushner (Univ. Michigan), Michael Lieberman (UC Berkeley), Yevgeny Raitses (Princeton Plasma Physics Laboratory)

Research frontier and importance of the scientific challenge.

Discharges with crossed electric and magnetic fields, or **E**x**B** discharges for short, enable plasma to form at reduced pressure-distance products because electrons are trapped, in a first approximation, and drift on a closed path. These discharges display remarkable self-organized structures in multiple dimensions, with transport coefficients that cannot be described by current theories. In spite of the prevalence of these time dependent, periodic, self-organized structures, quasi-stable operation can be achieved in limited parameter spaces. This limited stability, and the ability to produce plasma at relatively low pressure, has enabled numerous applications, ranging from widely used sputtering magnetrons for thin film deposition [1] to Hall thrusters for space propulsion [2-4] and micro-discharges with magnetic fields [5]. Of special interest is the emerging field of high power impulse magnetron sputtering (HiPIMS) [6-8], a new approach to ionized physical vapor deposition (PVD). These apparently disconnected fields of application share a common physics base. *The research frontier is to gain deeper understanding of the shared plasma physics that produces self-organized structures in **E**x**B** discharges across a large dynamic range of operating conditions, and their consequences on cross-field particle transport*. In addition to enabling a deeper understanding of this nearly unexplored area of plasma physics, deeper knowledge of the shared physics will enable scaling of **E**x**B** discharges, which will in turn stimulate advancements in applications.

Despite great variation in size, power and power pattern, geometrical design, operational pressure, and application, all of these discharges produce *low-temperature, weakly ionized plasma*. They are

distinct from fusion-related plasmas; they are non-uniform with localized regions of enhanced degree of ionization. All of these discharges exhibit a host of instabilities, and most pronounced are classes of instabilities associated with the motion of magnetized electrons in crossed fields (characteristic frequency usually in the MHz region) and ionization waves (characteristic frequency usually in the 100 kHz region, or even lower). The latter, the ionization instabilities, become visible through suitable plasma diagnostics such as fast camera imaging [6, 8], time-resolved spectroscopy [9], collecting probes [10], emissive probes [11], particle energy/mass spectrometers [10], diode-laser absorption spectroscopy [12], and various laser-induced fluorescence methods [13-15]. Discoveries of moving ionization patterns have been independently made in each of the very different physical devices (e.g., thrusters to plasma deposition reactors), with various descriptions, terminologies, interpretations, and strategies for utilization or suppression of the patterns. The observed phenomena can be regular (periodic) or chaotic in certain parameter regimes, or disappear when damped by collisions (Figs. 1-5).

The Grand Challenge is to develop a unified understanding of **E**x**B** discharges that connect apparently similar self-organization and instabilities observed over this very large dynamic range, exploring which of the underlying physical processes and principles are common and which are specific. What is the nature and driving forces of the instabilities? Can they be exploited in a controlled manner for selected applications? Assuming generalized conditions for the appearance of ionization instabilities can be identified, the challenge is to come to a generalized understanding of formation conditions, their boundaries, shape and structure of self-organized patterns, and the transition to chaos. It will be critical to better understand the electron energy distribution functions, which are expected to be dependent on space and time and be significantly different than a Maxwellian distribution when electrons are in equilibrium.

Once the conditions have been understood, the next step is to consider implications for particle transport because particle transport is most critical in all of the applications. Research so far has provided evidence for faster-than-Bohm transport, indicative of the role of instabilities which can locally deform the crossed field structure.

Since all of the discharge plasmas are non-uniform, the general class of **E**x**B** discharges also involves other drifts such as the gradient **B**-drift and higher order drifts. The role of the various drifts has not been sufficiently studied, and it is not even clear if the hydrodynamic-style distinction of different drift types is meaningful.

Approach to advancing the frontier, new research tools and capabilities required.

<u>a) Theory & simulation</u>

Computer simulation of these non-uniform plasmas is challenging as very different time scales are involved (ps to ms), and the plasma density can vary by several orders of magnitude. Current theoretical models are therefore based on strong (sometimes over-simplifying) assumptions, such as azimuthal symmetry, which a priori prohibits the study of instabilities in that dimension. For example, models using a conceptual structuring of the plasma in distinct regions [16] give some insight in the physics but this approach prohibits the capture of transport properties associated with instabilities.

Particle-in-Cell (PIC) codes are able to show the onset and evolution of instabilities but have encountered issues associated with locally high plasma density and dynamics, which is ultimately a 3D phenomenon. Therefore, approaches so far focus on the exploring the lowest plasma density, e.g. for low-power dc discharges [17]. Fluid-hydrodynamic codes have difficulty in addressing the conditions of these devices. The magnetic field configurations are highly non-uniform having magnitudes of a few Gauss to many kG in the region of interest, which then makes guiding center approximations inappropriate. For both PIC and fluid approaches, new modeling techniques will need to be developed, as total simulation times of ~100 μs represent a huge challenge, even when using high-performance-computing (HPC) platforms. That is, the problems will need HPC but likely they cannot be solved by simply using more processors – modern high-fidelity algorithms are required. Additionally, careful numerical analysis of modeling techniques will need to be developed and careful verification and validation of simulation results will need to be performed.

b) Experiment

As mentioned above, a range of plasma diagnostic tools have been applied so far, leading us to the importance of instabilities, self-organization, and faster-than-Bohm cross-field transport. Applying the essentially the same plasma diagnostic results to different types of **E**x**B** discharges could lead to the desired deeper insights as the common features can be parameterized and compared. Figures at the end of this White Paper illustrate this argument.

The most relevant and interesting physics happens is in relatively small regions of the plasma, as the plasma is non-uniform and dynamic. Therefore, plasma diagnostic tools need to have sufficient spatial and temporal resolution. For most of the applications, this implies spatial resolution of the order 1 mm; the necessary temporal resolution depends on whether one considers processes related to electrons only (nanoseconds or faster) or processes involving ions (e.g. ~1 μs).

In any case, as the critical physics is localized, the diagnostic tool needs to be able to "access" that particular region. Therefore, the best approach is to develop and apply improved non-intrusive plasma diagnostics, with resolution high enough to address spatial and temporal changes of the critical plasma parameters. Probes are useful in regions at some distance from where ionization instabilities develop, however, they disturb/affect the plasma when brought closely to the plasma's central **E**x**B** region. The use of antenna probes can mitigate this issue. In all cases, proper interpretation of probe signals need to take the influence of the magnetic field on the probe current into account, which calls for further development of probe theory in magnetic fields.

Non-invasive advanced optical and spectroscopic methods are of great interest, such as time and spectrally selective imaging [18]. "Active interrogation" has the greatest potential as one can be selective of what to probe. Among the active methods are diode-laser absorption spectroscopy [12], time-synchronized laser-induced fluorescence [15], and laser-induced fluorescence imaging [13], and Thomson scattering. These advanced methods require state-of-the-art laser and optical equipment, and develop their full potential when coupled with theoretical models and computer simulations.

Impact of this research on plasma science and related disciplines, potential for societal benefit.

Should this research be successful, a unified theory or understanding of **E**x**B** discharges will result in a new ability to control this important class of discharges. Each of the applications of **E**x**B** discharges has significant impact on its field, and has not been fully exploited for lack of this knowledge. For example,

- magnetron sputtering in its modern forms (pulsed, multiple frequencies, high peak power, with reactive gases) is key to many products, such as for displays and in microelectronics, wear and corrosion protective coatings on tools automotive and aerospace components, large area energy-savings coatings for low-emissivity and dynamic windows, just to name a few example of industrial use;
- Hall thrusters are mission-critical for deep-space exploration;
- Microdischarges can be enabling to some coatings, surface modifications, including modification of organic and biological specimens, and could be adapted as a miniaturized Hall thruster.

What is even more intriguing is the potential applications that might results from prevision control of **E**x**B** discharges. Plasma processing of extremely large areas (many meters on a side) at high rates is now limited by the ability to produce stable and uniform plasmas. Mastery of the self-organization and stability of **E**x**B** discharges will enable venturing into this new parameter space.

Figures

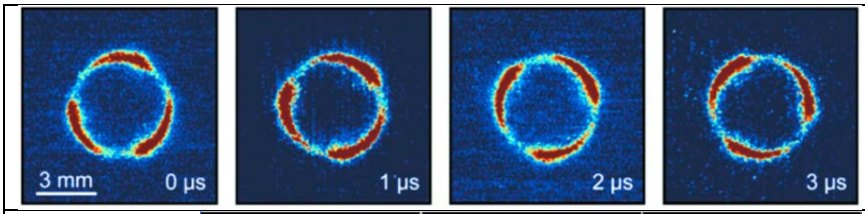

Fig. 1 Fast camera images of a magnetized micro-discharge, showing that the plasma contains a self-organized pattern, here moving in the –**E**x**B** direction (from [19]).

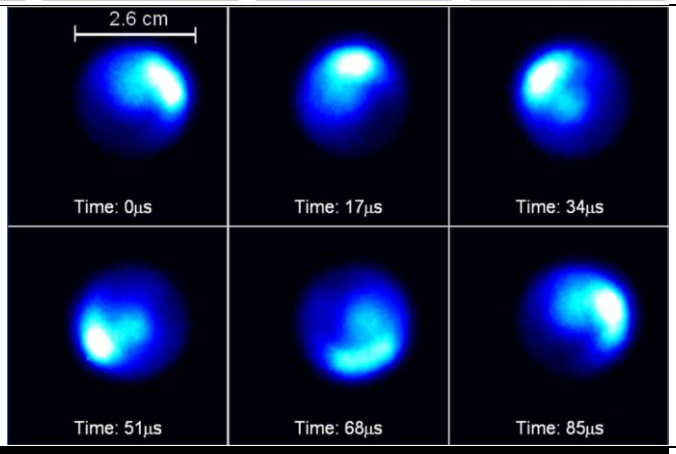

Fig. 2 Fast camera images of the rotating spoke instability in a 2.6 cm cylindrical Hall thruster. The 15–35 kHz spoke can be seen to rotate in the counterclockwise (**E**x**B**) direction. (from [2]).

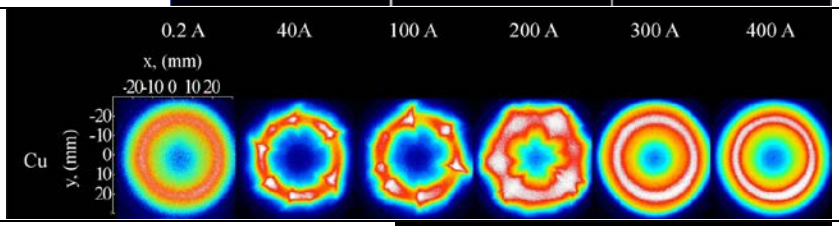

Fig. 3 Fast (3 ns) imaging of HiPIMS (3" magnetron). False color indicates intensity of light emitted (with different intensifier gain settings, from [20]).

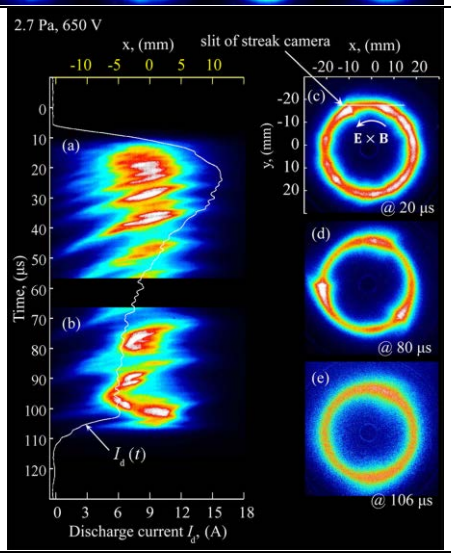

Fig. 4 Images of the ionization zone's propagation direction change in 2.7 Pa (Ar) and discharge voltage of 650 V. (a) and (b) streak camera image with a sweep time of 50 µs. Note the direction reversal of the ionization zone near the end of the pulse.
(c)–(e) Taken at different times within the pulse by an ICCD camera with exposure time 20 ns (from [21]).

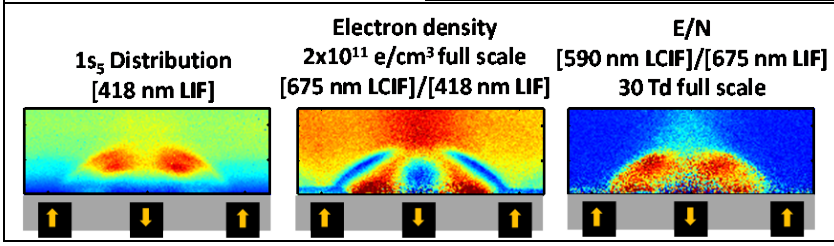

Fig. 5 Illustration of laser-induced fluorescence to measure plasma properties with high spatial, temporal, and spectral resolution (from [14]).

References

[1] G. Bräuer, B. Szyszka, M. Vergöhl, and R. Bandorf, "Magnetron sputtering - Milestones of 30 years," *Vacuum,* vol. 84, pp. 1354-1359, 2010.

[2] C. L. Ellison, Y. Raitses, and N. J. Fisch, "Cross-field electron transport induced by a rotating spoke in a cylindrical Hall thruster," *Phys. Plasmas,* vol. 19, p. 013503, 2012.

[3] E. Choueiri, "Plasma oscillations in Hall thrusters," *Phys. Plasmas,* vol. 8, p. 1411, 2001.

[4] E. Fernandez, M. K. Scharfe, C. A. Thomas, N. Gascon, and M. A. Cappelli, "Growth of resistive instabilities in E×B plasma discharge simulations," *Phys. Plasmas,* vol. 15, p. 012102, 2008.

[5] T. Ito and M. A. Cappelli, "High speed images of drift waves and turbulence in magnetized microplasmas," *IEEE Trans. Plasma Sci.,* vol. 36, pp. 1228-1229, 2008.

[6] A. Anders, P. Ni, and A. Rauch, "Drifting localization of ionization runaway: Unraveling the nature of anomalous transport in high power impulse magnetron sputtering," *J. Appl. Phys.,* vol. 111, p. 053304, 2012.

[7] J. T. Gudmundsson, N. Brenning, D. Lundin, and U. Helmersson, "High power impulse magnetron sputtering discharge," *J. Vac. Sci. Technol. A,* vol. 30, p. 030801, 2012.

[8] A. P. Ehiasarian, A. Hecimovic, T. de los Arcos, R. New, V. Schulz-von der Gathen, M. Böke, and J. Winter, "High power impulse magnetron sputtering discharges: instabilities and plasma self-organization," *Appl. Phys. Lett.,* vol. 100, p. 114101, 2012.

[9] M. Hala, N. Viau, O. Zabeida, J. E. Klemberg-Sapieha, and L. Martinu, "Dynamics of reactive high-power impulse magnetron sputtering discharge studied by time- and space-resolved optical emission spectroscopy and fast imaging," *J. Appl. Phys.,* vol. 107, p. 043305, 2010.

[10] M. Panjan, R. Franz, and A. Anders, "Azimuthally asymmetric particle fluxes in sputtering magnetrons, and their amplification by ionization zones in high power impulse magnetron sputtering," *Plasma Sources Sci. Technol.,* vol. 23, p. 025007, 2014.

[11] A. Rauch, R. Mendelsberg, J. M. Sanders, and A. Anders, "Plasma potential mapping of high power impulse magnetron sputtering discharges," *J. Appl. Phys.,* vol. 111, p. 083302, 2012.

[12] C. Vitelaru, D. Lundin, G. D. Stancu, N. Brenning, J. Bretagne, and T. Minea, "Argon metastables in HiPIMS: time-resolved tunable diode-laser diagnostics," *Plasma Sources Sci. Technol.,* vol. 21, p. 025010, 2012.

[13] E. V. Barnat and K. Frederickson, "Two-dimensional mapping of electron densities and temperatures using laser-collisional induced fluorescence," *Plasma Sources Sci. Technol.,* vol. 19, p. 055015, 2010.

[14] E. V. Barnat and B. R. Weatherford, "Two dimensional laser-collision induced fluorescence in low-pressure argon discharges," *Plasma Sources Sci. Technol.,* submitted for review, 2015.

[15] C. V. Young, A. Lucca Fabris, and M. A. Cappelli, "Ion dynamics in an E × B Hall plasma accelerator," *Appl. Phys. Lett.,* vol. 106, p. 044102, 2015.

[16] M. A. Raadu, I. Axnäs, J. T. Gudmundsson, C. Huo, and N. Brenning, "An ionization region model for high-power impulse magnetron sputtering discharges," *Plasma Sources Sci. Technol.,* vol. 20, p. 065007, 2011.

[17] A. Pflug, M. Siemers, T. Melzig, L. Schäfer, and G. Bräuer, "Simulation of linear magnetron discharges in 2D and 3D," *Surf. Coat. Technol.,* vol. 260, pp. 411-416, 2014.

[18] J. Andersson, P. Ni, and A. Anders, "Spectroscopic imaging of self-organization in high power impulse magnetron sputtering plasmas," *Appl. Phys. Lett.,* vol. 103, p. 054104, 2013.

[19] T. Ito and M. A. Cappelli, "Electrostatic probe disruption of drift waves in magnetized microdischarges," *Appl. Phys. Lett.,* vol. 94, pp. 211501-3, 2009.

[20] Y. Yang, K. Tanaka, J. Liu, and A. Anders, "Ion energies in high power impulse magnetron sputtering with and without localized ionization zones," *Appl. Phys. Lett.,* vol. 106, p. 124102, 2015.

[21] Y. Yang, J. Liu, L. Liu, and A. Anders, "Propagation direction reversal of ionization zones in the transition between high and low current magnetron sputtering," *Appl. Phys. Lett.,* vol. 105, p. 254101, 2014.