DOE Center for
Predictive Control of Plasma Kinetics:
Multi-Phase and Bounded Systems

5th Annual Meeting

May 15-16, 2014

University of Maryland, College Park, MD
Participating Institutions

We gratefully acknowledge the funding from the U.S. Department of Energy Office of Science Fusion Energy Sciences Program
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## Schedule

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#### 3:20 – 5:50 pm  
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8:25 – 8:50  Kentaro Hara (University of Michigan)
Direct Kinetic Simulation of the Discharge Plasma in a Hall Effect Thruster

8:50 – 9:15  JP Sheehan (University of Michigan)
Adiabatic Plasma Expansion in a Magnetic Nozzle

9:15 – 9:40  Ed Barnat (SNLA)
Diagnostics of Dynamic Plasma Discharges

9:40 – 10:05  Yevgeny Raitses (PPPL)
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10:05 – 10:25 am  Break

10:25 am – 12:00 pm  Group Discussion. Moderator: Mark J. Kushner
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2 Elliot Bartis (University of Maryland)
   *The Impact of Ambient Gas Chemistry on Lipopolysaccharide Deactivation and Polymer Modification by Plasma-Generated Radicals at Atmospheric Pressure*

3 Andrew Knoll (University of Maryland)
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4 James Mitrani (PPPL)
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5 Wei Tian (University of Michigan)
   *Long-term Interactions of Atmospheric Pressure Dielectric Barrier Discharge with Wet Tissues*

6 James Franek (West Virginia University)
   *Diagnostics for Fundamental Plasma Processes of Chemically Reactive Plasma Interacting with Surfaces with Emphasis on Near-atmospheric Systems*

7 Scott Keller (PPPL)
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8 Carlos Larriba-Andaluz (University of Minnesota)
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9 Vladimir Kolobov (CFDRC/University of Alabama at Huntsville)
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| 15  Peng Tian (University of Michigan)  
    *Controlling VUV Photon Fluxes in Pulsed Inductively Coupled Plasmas: Rare Gas Mixtures* |
| 16  Jeff Walker (West Virginia University)  
    *Non-Stationary Charging Dynamics of an Inhomogeneous Granule-Plasma Multi-phase System* |
| 17  Yiting Zhang (University of Michigan)  
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| 18  Timothy Sommerer (General Electric)  
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Abstracts: Oral Presentations

Introduction to the Annual Meeting: Subtleties in Control of LTPs
Mark J. Kushner
University of Michigan,
Electrical Engineering and Computer Science, Ann Arbor, MI 48109-2122 (mjkush@umich.edu)

The Department of Energy Center for the Predictive Control of Plasma Kinetics: Multi-Phase and Bounded Systems was established in the Fall of 2009 in response to recommendations of the National Research Council Plasma 2010 Decadal Study. The Plasma 2010 study emphasized the importance of low temperature plasmas (LTPs) to the economic well being and national security of the United States, and the high level of the fundamental science challenges facing the LTP field. These challenges were more definitively discussed at a DOE workshop whose report titled Low Temperature Plasmas: Not only the 4th State of Matter but All of Them proposed a roadmap to meet those challenges. The Center has been guided by that roadmap and so selected as its unifying theme the control of plasma kinetics.

Although the intellectual thrust of the Center remains well aligned with that unifying theme, during the 4+ years of the Center there has been a significant shift in the center-of-gravity of the field of LTPs from lower pressures to higher pressures, with motivating applications now including biotechnology. The fundamental research performed by the Center has continued to lead this evolution of the field. One of the outcomes of that research has been a deeper appreciation of the subtleties and dynamics of LTPs and the sophistication of techniques required to master those subtleties.

In this talk, a brief overview of the Center will be provided from these perspectives. The structure of the meeting and expected outcomes will be summarized. The subtleties encountered in controlling LTPs will be illustrated with examples from low and high pressure plasmas – phase control in dual frequency capacitively coupled plasmas and variability in the application of plasma jets to biomedical applications.
Plasma Diagnostics During Plasma Synthesis of Semiconductor Nanocrystals

Narula Bilik\textsuperscript{(a)}, Elijah Thimsen\textsuperscript{(a,b)}, Valery Godyak\textsuperscript{(c)}, Steven L. Girshick\textsuperscript{(a)}, Eray S. Aydil\textsuperscript{(b)} and Uwe Kortshagen\textsuperscript{(a)}

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Plasma synthesis of ≈1-20 nm semiconductor nanocrystals (NCs) is emerging as a high throughput, scalable and low-cost alternative to liquid based synthesis methods. Thin films comprised of these nanocrystals find applications ranging from solar cells to light emitting diodes. Plasmas offer several advantages. First, exothermic surface recombination reactions heat the NCs to temperatures hundreds of Kelvin above the gas temperature and enable the synthesis of high melting point materials. Second, plasma synthesis is free of solvents and surfactants. Third, doping and purity control is easier in the gas phase than in the liquid phase. Finally, plasma processes are higher throughput and continuous whereas most liquid-based methods are batch. While liquid-based synthesis is widely used and researched, plasma synthesis of nanocrystals is in its infancy. A combination of plasma diagnostics and NC characterization is needed to elucidate the key factors that control the NC and plasma properties.

Towards this end, we are conducting Langmuir probe measurements in plasmas containing NCs. These measurements are particularly challenging because probes are coated rapidly. We solve this problem using a solenoid actuator-controlled shield that exposes the probe wire to the plasma momentarily and only during the current-voltage sweep. Using this approach we have successfully measured the electron energy probability distribution function (EEPF) in a dusty plasma (silicon nanoparticles in SiH\textsubscript{4}/Ar discharge) under various conditions for the first time. For example, Figure 1 shows the EEPF in an initially pristine Ar plasma, after the injection of 5% SiH\textsubscript{4} in Ar to form Si nanoparticles (dusty plasma) and after the Si nanoparticles have been purged to return the Ar plasma to its pristine state. The dusty plasma EEPF, when compared to that of the pristine Ar plasma, shows an increase in high-energy electrons (>7eV) and depletion of low-energy electrons (<7eV), leading to an increased average electron temperature.

Finally, we will summarize the synthesis of a variety of binary semiconductor nanocrystals from metalorganic precursors including ZnS, Cu\textsubscript{2}S, SnS and ZnO. Optical emission spectroscopy was used during synthesis to obtain insight into the plasma chemistry.

![Figure 1 – EEPFs of pristine argon plasma at 80 mTorr and 40 W before Si NC (dust) formation (red), after injection of 5% SiH\textsubscript{4} in Ar to form dust (green) and after purging the dust (blue).](image)
Designing Nanocrystal Plasma Synthesis at Atmospheric Pressures

Nicolaas J. Kramer\(^{(a)}\), Eray S. Aydil\(^{(b)}\) and Uwe R. Kortshagen\(^{(a)}\)

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Plasma growth and crystallization of nanoparticles is an exciting new frontier both for plasma science as well as materials research. The unique ability of plasmas to produce well-defined nanocrystals of high melting point materials through selective nanoparticle heating while the gas remains at room temperature has already enabled the synthesis of new classes of nanomaterials. To date, the mechanisms of nanoparticle charging and heating in nonthermal plasmas have been studied and understood to some extent for low pressure plasmas. However, in order to make plasma synthesis in nanodusty plasmas attractive for industrial applications, moving the field of dusty plasmas to atmospheric pressures is crucial.

The fundamental processes of nanoparticle charging and heating are significantly different at atmospheric pressure compared to low pressures. Charging is determined through hydrodynamic, mobility driven collection of ions by the nanoparticles rather than by orbital motion at low pressures. Nanoparticle heating reactions have to compete with nanoparticle cooling through convection/conduction to the neutral gas that is about 100-1000 times faster than at low pressure.

In this presentation we present results of a nanoparticle heating and charging model that is applicable over a wide range of pressures. Nanoparticle heating reactions such as electron-ion recombination and energetic surface reactions are treated stochastically through a Monte Carlo approach. Nanoparticle cooling through conduction/convection is modeled through a continuum model. The model indicates that the relation between the characteristic time for nanoparticle heating reactions and the nanoparticle temperature relaxation time is essential for the nanoparticle heating mechanism. At low pressure, several heating reactions often occur within one temperature relaxation time, leading to the high nanoparticle temperatures that are required for nanoparticle crystallization. At atmospheric pressure, the nanoparticle temperature relaxation time is much shorter and successive reactions within one temperature relaxation time are much less likely. Hence the nanoparticle temperature on average remains much closer to the gas temperature. Based on this model, we predict that the formation of nanocrystals at atmospheric pressure requires significantly higher plasma densities than at low pressure. We suggest design principles for nanocrystal synthesis at atmospheric pressures.

Figure 1 – Nanoparticle temperature ranges as function of pressure and ion density. The dashed line indicates the crystallization temperature of 5 nm Si particles.
Numerical Modeling of Nanodusty Plasmas

S. L. Girshick\(^{(a)}\), C. Larriba-Andaluz\(^{(a)}\), R. Le Picard\(^{(a)}\), S.-H. Song\(^{(b)}\) and M. J. Kushner\(^{(b)}\)

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\(^{(b)}\) Department of Electrical Engineering and Computer Science, University of Michigan

Nanodusty plasmas are plasmas in which nanoparticles nucleate and grow. Plasmas have several potential advantages for synthesizing nanoparticles for applications that include solar cells, optoelectronics, catalysis, and biomedical. On the other hand, unwanted formation of nanoparticles represents an important source of contamination in semiconductor processing.

In a collaboration between the University of Minnesota and the University of Michigan, we are developing numerical models of several types of nanodusty plasmas, with a focus on synthesis of silicon nanoparticles in silane-containing plasmas. Systems modeled include a parallel-plate capacitively-coupled RF system at pressures in the 100 mTorr range (both continuous and pulsed), a flow-through cylindrical tube with RF ring electrodes and pressure around 2 Torr [1], and, in proposed work, atmospheric-pressure nanodusty plasmas in transverse flow geometry.

Nanodusty plasmas typically involve a strong coupling between the nanoparticle cloud and the plasma, presenting severe challenges for computational modeling. In this work the sectional aerosol model developed at Minnesota [2,3] is integrated into Michigan’s Hybrid Plasma Equipment Model [4] for modeling plasma flow through a cylindrical tube, and the non-PDPSIM code [5] for the proposed work on atmospheric-pressure dusty plasmas. The work on pulsed parallel-plate RF plasmas involves a collaboration with Ecole Polytechnique, Palaiseau, France, with the objective of controlling the sizes and fluxes of energetic charged particles to a substrate during growth of epitaxial silicon films. All of these projects involve experimental validation in collaboration with others.

References

Figure 1 – Parallel-plate capacitively-coupled RF dusty plasma with pulsing of applied voltage. Top: negative nanoparticles are trapped in plasma, positive particles accelerated to substrate by sheath potential and, during off phase, DC negative bias. Bottom: simulation results for fraction of nanoparticle flux to substrate that is positive (without biasing), for 100-Hz pulsing with 50% duty cycle.
The Magnetized Dusty Plasma Experiment: A Status Report

Edward Thomas, Jr.\textsuperscript{(a)}, Uwe Konopka\textsuperscript{(a)}, Ami DuBois\textsuperscript{(a)}, Brian Lynch\textsuperscript{(a)}, Stephen Adams\textsuperscript{(a)}, Spencer LeBlanc\textsuperscript{(a)}, Darrick Artis\textsuperscript{(a)}, Robert L. Merlino\textsuperscript{(b)} and Marlene Rosenberg\textsuperscript{(c)}

\textsuperscript{(a)} Physics Department, Auburn University (etjr@auburn.edu)
\textsuperscript{(b)} Department of Physics and Astronomy, The University of Iowa
\textsuperscript{(c)} Department of Electrical and Computer, University of California – San Diego

Over the last two and one-half years, the Magnetized Dusty Plasma Experiment (MDPX) has been under development at Auburn University [1]. The MDPX device consists of a four-coil, superconducting magnet system that can produce magnetic field strengths in excess of 4 Tesla in the bore. In the bore, an 18” diameter, octagonal vacuum chamber will be used to study strongly magnetized plasmas and dusty plasmas. The primary missions of the MDPX device are to: (1) investigate the structural, thermal, charging, and collective properties of a plasma as the electrons, ions, and finally charged microparticles become magnetized; (2) study the evolution of a dusty plasma containing magnetic particles (paramagnetic, super-paramagnetic, or ferromagnetic particles) in the presence of uniform and non-uniform magnetic fields; (3) explore the fundamental properties of strongly magnetized plasmas (“i.e., dust-free” plasmas).

At the end of February, 2014, the MDPX magnet was delivered to Auburn University. By mid-April, 2014, the first magnetic field operations and first plasma operations of the MDPX device began. This brief presentation will discuss the initial characterization of the magnetic field structure in the MDPX device and will report on initial plasma operations.

References

Figure 1 – Photo of the cryostat of the MDPX device. The gap in the center will contain the vacuum chamber. The overall height of the black cryostat structure is ~120 inches.

Figure 2 – Photograph of the MDPX vacuum chamber on a test stand in the laboratory – prior to the delivery of the magnet.
Low-temperature Plasma-surface Interactions: Atmospheric Pressure Plasma Treatment of Polymers/biomolecules and Atomic Layer Etching of SiO2/Si

E.A.J. Bartis(a), D. Metzler(a), A. Knoll(a), P. Luan(a), D.B. Graves(b), W.R. Lempert(c), I. Adamovich(c), V. Godyak(d), M.J. Kushner(d), and G.S. Oehrlein(a)

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Atmospheric pressure low temperature plasma sources have attracted substantial attention since plasma-generated reactive species can deactivate bacteria/biomolecules or modify surfaces. Lipopolysaccharide (LPS), a toxic biomolecule, and a 193 nm photoresist (PR) polymer were exposed to an Ar atmospheric pressure plasma jet (APPJ) with small admixtures of O2, N2, and H2O. LPS biological activity was monitored by an immunosorbent assay and PR etch rates were measured by in situ ellipsometry. For both materials, the surface chemistry was measured by x-ray photoelectron spectroscopy (XPS). The APPJ is mounted inside a vacuum chamber that can be evacuated and then refilled with a controlled environment. We regulate the interaction of the plasma with the ambient by confining the plasma inside an alumina tube or allowing it to extend out of the tube as a plume. When the Ar/O2 plume is exposed to the environment, surface modifications such as oxygen uptake and formation of surface-bound NOx reflect the ambient N2 concentrations in N2/Ar environments. For instance, XPS analysis of a photoresist polymer without nitrogen shows formation of surface NOx species after Ar/1% O2 APPJ treatment, demonstrating that NOx surface species (tentatively identified as NO3) are created by plasma-environment interactions (see Fig. 1). This results from the plasma exciting ambient N2, which quenches reactive oxygen species that would otherwise modify the films. Consistent with this quenching role, N2 addition to O2/Ar feed gas results in decreased densities of reactive oxygen species (e.g. atomic O emission, O3) and impact on bioactivity of biomolecules. Material etch rates and surface modifications measured by real-time ellipsometry for different APPJ exposure conditions/environmental interactions will also be reported.

Atomic layer etching of SiO2 is enabled by low temperature Ar plasma in conjunction with periodic injection of a defined number of C4F8 molecules and synchronized plasma-based Ar+ ion bombardment. Precise management of C4F8 supply allows control of the deposited fluorocarbon (FC) layer thickness in the 1 to several Ångstrom range. For low energy Ar+ ion bombardment conditions, the physical sputter rate of SiO2 vanishes whereas when FC reactants are present at the SiO2 surface etching can be initiated. The impact of deposited FC layer thickness and ion energy on SiO2 etching and etch selectivity relative to Si is examined and quantified. Additionally, we investigated surface chemistry at various stages of the cyclic etching with XPS, and plasma potential, electron temperature, and electron density during cyclic exposures using a Langmuir probe.

The authors gratefully acknowledge financial support from US Department of Energy (DE-SC0001939 and DE-SC0005105) and the National Science Foundation (PHY-1004256 and CBET-1134273). We also thank R.L. Bruce, S. Engelmann, and E.A. Joseph of IBM T.J. Watson Research Center, Yorktown Heights for their collaboration on atomic layer etching.
From Nonlocal Kinetic Theory to Practical Applications

I.D. Kaganovich(a), D. Sydorenko(b), A.V. Khrabrov(a), Y. Raitses(a), M.D. Campanelli(d), H. Wang(a), E. Tokluoglu(a), P. Ventzek(c), L. Chen(c), V.I. Demidov(d), M.E. Koepke(d), A.S. Mustafaev(e)

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The purpose of the talk is to describe recent advances in nonlocal electron kinetics in low-pressure plasmas enabled by the DOE Center for Control of Plasma Kinetics. Low-pressure discharges are widely used in industry as the main plasma sources for many applications including plasma processing, discharge lighting, plasma propulsion, particle beam sources and nanotechnology. Being partially-ionized, bounded, and weakly-collisional, the plasmas in these discharges demonstrate nonlocal electron kinetic effects, nonlinear processes in the sheaths, beam-plasma interaction, collisionless electron heating, etc. Such plasmas often have a non-Maxwellian electron velocity distribution function. Recently Physics of Plasmas published special topic of collected papers dedicated to “Electron kinetic effects in low temperature plasmas” in memory of the pioneer and leader of this field, Professor Lev D. Tsendin [1]. The plethora of kinetic processes supporting the non-equilibrium plasma state is an invaluable tool, which can be used to adjust plasma parameters to the specific needs of a particular plasma application. We report on recent advances in nonlocal electron kinetics in low-pressure plasmas where a non-Maxwellian electron velocity distribution function was “designed” for a specific application: in dc discharges with auxiliary biased electrodes for plasma control [2], hybrid DC/RF unmagnetized [3] and magnetized plasma sources [4], and Hall thruster discharges [5]. We show using specific examples that this progress was made possible by synergy between full-scale particle-in-cell simulations, analytical models, and experiments. Initial “academic” studies paved the way to understanding of modern plasma devices that are being developed for future plasma technology. One example is so-called non-ambipolar electron plasma, where an electron beam is extracted through a small aperture [6]. Our previous studies of extraction system [2] and collective interaction of electron beam with the plasma [3] aides understanding and optimization of this device [6]. Another example is modeling of high power plasma switch for electric grid system [7]. Developed analytical and simulation tools are now being used to provide optimization strategy for the device.

In summary, the Center funding allowed us to develop set of valuable analytical and simulation tools that are now being used in industry to increase US competitiveness.

References
Mechanisms of Electron Acceleration by Two-stream Instability in DC Discharge
D. Sydorenko\textsuperscript{(a)}, I. D. Kaganovich\textsuperscript{(b)}, A. V. Khrabrov\textsuperscript{(b)}, P. L. G. Ventzek\textsuperscript{(c)}, and L. Chen\textsuperscript{(c)}

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The interaction of an electron beam with plasma is of particular importance for hybrid DC/RF coupled plasma sources used in plasma processing [1,2]. Electron acceleration by high-frequency waves may explain the low-energy peak in the electron energy distribution function measured in plasma processing devices [1]. We continued our studies of mechanisms of the collisionless electron heating in a hybrid RF-DC plasma source using the particle-in-cell code EDIPIC [3]. Different emission currents and neutral argon gas pressures are considered. The plasma size, density, dc voltage, and neutral gas pressure are close to the parameters of a commercial RF-DC discharge used for material processing. The beam electrons are emitted from the cathode and accelerated by the dc voltage applied to the system. The beam excites electron plasma waves through the two-stream instability. The plasma density profile is maximal in the middle and decays towards the plasma edges. It is found that at higher pressures, when the mean free path of bulk electrons is less than the plasma size, the instability does not develop if the emission current is below some threshold. The instability reappears for the low current if the neutral gas density decreases. The growth rate in finite size plasma is significantly less than that in infinite plasma. When the instability occurs, a global mode is excited with uniform frequency in the entire discharge as evident in Fig.1. The long plasma waves resonant with the beam are converted to much shorter plasma waves in the areas where the plasma density has a gradient as shown in Fig.1. The short waves have relatively slow phase speed comparable with that of thermal electrons and thus this transformation from the long wave to short waves due plasma inhomogeneity may be efficient mechanism for acceleration of plasma bulk electrons.

References
Interest in pulsed plasmas has re-emerged in recent years. Among the advantages, the control of the electron energy distribution (EED) provided by pulsing allows more control over the neutral and ion plasma chemistry. There is also interest in obtaining nearly monoenergetic ion energy distributions (IED) for improved control of ion-bombardment-stimulated surface processes, especially near an energy threshold for such processes. In ICPs, the IED is broadened during the power-on period by capacitive-coupling by the high-voltage end of the coil, imposing an RF potential on the DC plasma potential. This can be eliminated with a Faraday shield, but with electronegative gases it is not possible to ignite pulsed ICPs, since electron density rapidly decays when power is off, and re-ignition requires large electric fields produced by high-voltage capacitive coupling. Motivated by this problem, we have explored a “tandem” plasma system, where a continuous (upper) ICP is injected through a grid into a pulsed (lower) ICP. The expectation was that the injected plasma would provide a sufficient electron number density to allow the pulsed ICP to re-ignite rapidly when power is pulsed on.

Time-resolved Langmuir probe and optical emission spectroscopy (OES) were carried out mostly in pure Cl$_2$ plasmas. OES measurements are shown in Fig. 1A as a function of time and duty cycle for one 1000 µs cycle. With a duty cycle of 20%, emission rises sharply with plasma re-ignition and continues to rise slowly until dropping rapidly as power is extinguished. As the power-on time increases, a delay in plasma re-ignition begins to set in. With an 80% duty cycle, the delay is about 140 µs, and at 99% duty cycle it is 325 µs. Only when the duty cycle is >99.6% does the delay time become comparable to the power-off time. Delay times as a function of duty cycle are shown in Fig. 1B. Langmuir probe measurements (not shown) mirror these trends. Electron densities ($n_e$) exhibit similar delays at larger duty cycles before increasing to steady values. $n_e$ decays rapidly after power is extinguished; more rapidly than positive ion densities as Cl$^-$ forms. Re-ignition occurs roughly when the electron density decays to the level provided by the upper CW ICP. A global model was developed which predicts many of these trends. The delay appears to be due to the fact that the plasma density loss rate exceeds the formation rate early in the off period. The formation rate slows because the rapid drop in $n_e$, due to attachment, causes a drop in efficiency in the matching network. When the loss rate becomes less than the formation rate late in the off period, $n_e$ begins to rise, which improves the matching network efficiency, causing $n_e$ to rise faster, leading to re-ignition. Other electronegative plasmas (CF$_4$/O$_2$, SF$_6$) exhibit similar trends; Ar does not!

![Figure 1 - A) Cl emission intensity vs. time for a 5 mTorr ICP. B) Delay times for the ignition of plasma vs. power-on duty cycle.](image-url)
Strong secondary, thermionic and photon-induced electron emission occurs in many plasma systems. We show with theory and simulations some new emissive sheath phenomena that can fundamentally change the plasma-surface interaction under certain conditions.

**Emission can make sheaths positive** – When the emission is too strong, classical Debye sheaths cannot maintain zero current (Fig. top left). Past theories predict a space charge limited (SCL) sheath forms, where they still assume ions flow to the wall via Bohm’s criterion.[1] But another sheath is possible, the “inverse sheath” where ions are repelled.[2] In simulation works with SCL sheaths, ions were accelerated towards the wall by artificial “source sheaths”. [3] We found if a simulated plasma is produced realistically by volumetric ionization, inverse sheaths form. Ions are trapped and there is no charge loss or sputtering at the wall. A recent experiment showed inverse sheath formation![4]

**Emission can change the structure of the plasma interior** [2] – SCL sheath theories assume that a conventional presheath accelerates ions to the Bohm velocity at the edge.[1] But in the inverse sheath state, the whole plasma structure changes because no gradients are needed to accelerate ions. The ion flow velocity is exactly zero throughout the plasma interior.

**Secondary emission can cause instabilities** [5] – A general theory describes various conditions under which secondary emission causes sheath instabilities. Instabilities can modulate particle and energy fluxes, prevent steady state from being reached, and enhance cross-B transport. One example is the “weakly-confined electron instability” predicted to occur under high voltage regimes in Hall thrusters. Instabilities are not captured in time-independent sheath theories.

**Emitting sheaths can interact with each other** [6] – Most models of emission treat a plasma contacting one wall. But in weakly-collisional plasmas, emitted electrons can transit the plasma gap and reach the other wall. In asymmetric systems, transiting “beams” do not cancel. A complex flux balance problem arises where the sheath potentials are coupled by the electron exchange.

**References**
Anticipated Process of Gathering Input from the Science, Engineering, and Technology Community for FESAC’s Priorities Assessment and Budget Scenarios

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The DOE’s Fusion Energy Sciences (FES) Advisory Committee (FESAC) is charged to assess the priorities among continuing and potential new scientific, engineering, and technical research program investments within FES required to ensure that the U.S. is in a position to exert long-term leadership roles within and among each of the three areas in FES’s newly structured program: (1) the science of prediction and control of burning plasmas ranging from the strongly-driven state to the self-heated state, (2) the science of fusion plasmas, plasma-material interactions, engineering and materials physics modeling and experimental validation, and fusion nuclear science approaching and beyond ITER-relevant heat fluxes neutron fluences, and pulse lengths, and (3) the study of laboratory plasmas and the high-energy-density state relevant to astrophysical phenomena, the development of advanced measurement validation, and the science of plasma control important to industrial applications. A fourth area, establishing the scientific basis for attractive, robust control of the self-heated, burning plasma state, uses ITER as the keystone, is not so focused on domestic capabilities, and is not emphasized in this charge.

So that FES can formulate the FES strategic plan required by the FY2014 Omnibus Appropriations Act by mid-January 2013, the DOE Office of Science (DOE-SC) asks FESAC (a) to prioritize between the FES-defined program elements, (b) to include FESAC views on new facilities, new research initiatives, and facility closures, (c) to establish a scientific basis for advancing fusion nuclear science, (d) to assess potential for strengthened or new partnerships with other federal agencies and international research programs that foster opportunities otherwise unavailable to FES-supported scientists, and (e) to make use of prior studies and reports.[1-5]

I will outline the anticipated process whereby the basic, applied, and fusion research communities will be encouraged to provide input to the FESAC Strategic Planning panel.

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Air Plasma Creation of Nitrogen Oxides

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Atmospheric-pressure air plasmas are promising sources of reactive nitrogen oxide species for the emerging field of plasma biotechnology and related fields. We describe quantitative gas-phase measurements of plasma chemistry via infrared and ultraviolet absorption spectroscopy in confined volumes. \cite{1,2} We quantify the gaseous concentrations of nitrogen oxides (NO and NO\textsubscript{2}, or NO\textsubscript{x}), as well as ozone (O\textsubscript{3}) throughout various operating regimes. When power density is below about 0.1 W/cm\textsuperscript{2}, the discharge creates ozone; above about 0.2 W/cm\textsuperscript{2}, the discharge creates NO, NO\textsubscript{2} and N\textsubscript{2}O in approximately equal concentrations, increasing to about 1400 ppm (~ 10\textsuperscript{17} cm\textsuperscript{-3}) after about 10 minutes. The most rapid rate of creation of NO\textsubscript{x} species, however, occurs in spark-like discharges. These discharges can create NO\textsubscript{x} at rates about one order of magnitude greater. The relative rates of creation of NO and NO\textsubscript{2} in the air spark-like discharges can be controlled to some extent by the form of the applied voltage. A rapid increase in applied voltage followed by breakdown and immediately turning off the discharge tends to create more NO. Prolonged ‘glow-like’ discharges create more NO\textsubscript{2}. We have correlated the gas phase species concentrations vs. time in the enclosed volume with the (dried film surface) bacterial killing efficiency. The NO\textsubscript{x} mode appears more effective in killing bacteria dried on surfaces.

Nitrogen oxides are known to exert important biochemical effects in many systems, and have been investigated as key agents in therapeutic applications such as cancer therapy, blood flow modification and wound healing. \cite{3} When these species enter water, they form nitrite and nitrate anions (NO\textsubscript{2}\textsuperscript{-}, NO\textsubscript{3}\textsuperscript{-}). Reactive oxygen will often generate hydrogen peroxide (H\textsubscript{2}O\textsubscript{2}). These species react to form peroxynitrite (ONOO\textsuperscript{-}), another important bioactive molecule. \cite{4} Creating and controlling the reactive nitrogen species from air plasma is a crucially important aspect of atmospheric pressure plasma applications in a wide range of applications, ranging from food and surface disinfection, agricultural applications, device and instrument sterilization, and virtually all biomedical applications.

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Non-Equilibrium Atmospheric Pressure Plasma Jet: Reactive Chemistry, Transport and Interaction with Liquids

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Non-equilibrium atmospheric pressure plasma jets (APPJs) have received a lot of attention in last decade due to their huge potential for biomedical and material applications. As they are relatively inexpensive to implement, every group around the world has its own plasma jet. However the large variety of jets makes a comparison of results between different groups extremely complex. This is enhanced by the limited results available in literature on the plasma properties of APPJs. APPJs are typically operated in argon or helium in the humid air environment of the lab. This is additionally enhanced by the high complexity of non-equilibrium physical and chemical processes occurring in these plasmas, plasma-flow interaction (and related air diffusion in the effluent) and the interaction of the plasma with substrates and liquids.

During the last years in our group at the Eindhoven University of Technology, we have characterized a RF driven APPJ in great detail. The characterization includes electrical measurements, imaging, optical emission spectroscopy, (two photon enhanced) laser induced fluorescence, Thomson scattering, Rayleigh scattering, Raman scattering and mass spectrometry. This led to a detailed knowledge of the electron density, electron temperature, gas temperature, NO, O, OH, O\(_3\) densities, ionic species and air concentrations in the plasma effluent [1-5].

This well characterized jet has been used to investigate the reactive chemistry, its transport in the gas phase and transfer to the liquid phase. The focus has been on the NO and O related chemistry and the transfer of reactivity in the form of reactive nitrogen species, O\(_3\) and H\(_2\)O\(_2\) to the liquid phase. It is shown that VUV radiation and gas flow is involved in the reactivity transfer from the gas to the liquid phase. The coupling of the plasma to a substrate has an important effect on the power consumption and the morphology of the discharges with initial indications of a significant change in plasma induced liquid phase chemistry.

References
Modeling of High-Power Gas Switch for Electric Grid System

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There has been recent interest in utilizing gas switches in high-power AC/DC conversion for the purpose of power transmission over long distances. These devices would be based on a glow discharge with magnetically insulated cold cathode [1]. Their operation is similar to sputtering magnetrons [2,3], but at much higher pressures (0.1 to 1 Torr) in order to achieve high current densities.

We present results of numerical (the particle-in-cell code EDIPIC 1d3v PIC [4]) and analytical investigation of a gas switch in the conduction phase. The important properties of the high-pressure magnetron discharge are a very narrow cathode sheath and a considerable voltage drop in the magnetized pre-sheath where most of the ionization takes place due to Joule heating.

References
Hybrid Global Model Simulations of He/N₂ and He/H₂O Atmospheric Pressure Capacitive Discharges

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Atmospheric pressure radio-frequency (rf) capacitive discharges are of interest for materials processing and bio-medical applications. We have used 1D particle-in-cell (PIC) simulations of an atmospheric He/0.1%N₂ discharge with simplified chemistry to guide the development of a hybrid analytical/numerical global model that includes electron multiplication and two classes of electrons: “hot” electrons associated with the sheaths, and “warm” electrons associated with the bulk [1]. The fields and electron powers are found analytically to determine the time-varying hot and warm temperatures and the rate coefficients, and the particle balance equations are integrated numerically to determine the species densities. The model and PIC results show reasonable agreement and indicate a transition from a low power \(\alpha\)-mode having relatively high bulk electron temperature \(T_e\) to a high power \(\gamma\)-mode with a low \(T_e\). The transition is accompanied by an increase in density and a decrease in sheath widths.

Water is a trace gas of some interest for bio-medical applications, particularly since it may arise from contact with a subject who is being treated. We use the hybrid global model to simulate a chemically complex, bounded He/H₂O atmospheric pressure discharge [2], including 148 volume reactions among 43 species, and including clusters up to H₁₉O⁹⁺ [3]. The discharge is embedded in a larger volume, in which the trace gas fraction is controlled, leading to depletion of water within the discharge and diffusive flows of reaction products to the walls. For a planar discharge with a 1 cm electrode radius and a 0.5 mm gap driven at 13.56 MHz, we determine the depletion and diffusion effects and the \(\alpha\) to \(\gamma\) transition for secondary emission \(\gamma_{se}=0.25\) over a range of rf currents and external H₂O concentrations. Each simulation takes about 2 minutes on a medium size laptop computer, allowing exploration of a large input parameter space.

References
Characterization and Modeling of Transient Ionization Wave Discharges

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Nanosecond pulse discharges in point-to-point and near-surface ionization wave geometries have been studied experimentally and theoretically in a number of gases and gas mixtures (He, H₂, He-H₂, air, air-H₂, Ar-O₂-H₂, N₂-H₂O, and Ar-H₂O). The principal objective of these studies is to elucidate kinetic mechanisms of discharge development and plasma chemical reactions involved. Time-resolved electron density and electron temperature in nanosecond pulse, “diffuse filament” discharges sustained between two spherical electrodes in He and He-H₂ mixtures at P=100-200 torr have been measured by Thomson scattering. Peak electron number density and peak electron temperature are \( n_e \approx 3.5 \cdot 10^{15} \text{ cm}^{-3} \) and \( T_e \approx 4 \text{ eV} \), respectively. The kinetic model predictions agree well with the temporal trends detected in the experiment (such as rapid initial rise of electron temperature and electron density during the discharge pulse, and gradual decay in the afterglow), although peak electron temperature and electron density values during the pulse are somewhat overpredicted (see Fig. 1).

Time-resolved electric field in a nanosecond pulse, plane-to-plane, high-pressure discharge in H₂ has been measured by psec CARS/4-wave mixing. The results have been compared with kinetic modeling calculations, showing good agreement. It is anticipated that electric field during nanosecond pulse breakdown in a diffuse, high-pressure, dielectric barrier discharge will be measured with sub-nsec resolution, within the next few weeks. Time-resolved and spatially resolved temperature was measured in “diffuse” filament nanosecond pulse discharges in air and hydrogen-air mixtures, using purely rotational CARS spectroscopy. The results demonstrate that temperature rise in these transient plasmas occurs in two stages: “rapid heating”, during collisional quenching of excited electronic states of nitrogen, and “slow heating”, caused by vibrational relaxation of nitrogen in the ground electronic state, in good agreement with kinetic modeling predictions. Time-resolved, 1-D and 2-D distributions of temperature and absolute number densities of radical species (OH and H) have been measured by OH LIF, H atom TALIF, and Rayleigh scattering in two nanosecond pulse discharge configurations, (a) “diffuse filament” discharge in H₂-O₂-Ar mixtures, and (b) near-surface discharge sustained at the liquid water / water vapor interface. Further insight into dynamics of a surface ionization wave discharge developing over liquid-vapor interface is obtained by nsec gate plasma emission imaging and FTIR spectroscopy. FTIR absorption spectra of products accumulated in surface ionization wave discharges at liquid water / water vapor interface and liquid butanol / butanol vapor interface have shown significant NO₂ accumulation, as well as conversion of butanol into a number of lighter hydrocarbon species, demonstrating significant potential for surface plasma processing of evaporating reactants at room temperature.

![Figure 1 - Electron density and electron temperature during a nanosecond pulse discharge in He at 200 torr, plotted in two different time scales. He, P=200 torr.](image)
Correlating Post-Transient Metastable-Atom Density, Reduced Electric Field, and Electron Energy to 420nm/419nm Ratio in a Pulsed Argon Discharge Column *

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Direct measurement of the intensities of the Ar 420.1nm and 419.8nm spectral lines, independent measurement of metastable-atom density, electron density, and electron temperature, inferred measurement of reduced electric field, and assumptions of the shape of the electron energy distribution in a 1-torr argon discharge are correlated within the extended-corona-model framework applied to the late stage of a pulsed discharge-tube plasma to determine the reaction rate constants $k_{ij}$ that are used to predict the 420.1nm to 419.8nm line ratio [1]. The model predicts post-transient metastable density. The diagnostic use of optical emission spectroscopy (OES) to interpret experimental conditions for $t>20 \mu s$ into the pulsed discharge is discussed.

The Ar 420.1nm and 419.8nm spectral line intensities were measured using a 0.5m Jobin Yvon Spectrometer, with a 2400 lines/mm diffraction grating and an Andor iStar ICCD camera. The density of 1s5 argon metastable atoms in the discharge was measured by laser diode absorption spectroscopy using a tunable diode laser. Electron density and electron temperature measurements were obtained using microwave techniques [2]. Plasma current was measured and combined with the results of Pack, et al. [3] to infer the local value of reduced electric field ($E/n$). The value of reduced electric field and the results of Adams et al. [1] were combined to infer the values of the reaction rate constants.

Insight into plasma conditions and their time dependence is afforded by invoking plausible assumptions regarding the shape of the electron energy distribution function (EEDF) which evolves and may or may not be consistent with invoked assumptions at all times. The next step is to validate assumptions regarding the EEDF shape in the discharge.

References

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Controlling Plasma Properties in DC Discharges with Two Anodes

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In a dc discharge plasma with two anodes, the second anode can be effectively used for robust control of plasma properties. The device operation is based on nonlocal electron kinetics; therefore it is called nonlocal plasma (NLP) technology in Ref. [1]. The device has a number of advantages compared to a regular device with one anode; for example, discharge instabilities are effectively suppressed when the second anode is used [2]. The discharge device with thermionic emission from cathode has been studied in this work (see, figure 1). In the device the main anode is placed between the cathode and the other auxiliary anode and has a circular opening for passing electron current from the cathode to the second anode. This configuration is also relevant to novel electron beam extraction system of Ref. [3].

It is experimentally demonstrated that a plasma may exhibit a sharp transition between two quasi-stable conditions, dependent on the cathode-electron current collected by the auxiliary anode through an aperture, i.e., hole, in the main anode. In the first regime, a bright ball-shaped glow is formed in front of the opening on the plasma side of the main anode. A glow is caused by electron acceleration in a potential drop of 10-15 eV formed in plasma near opening. The second regime is characterized by a uniform potential profile and uniform glow in plasma and an absence of the ball-shaped glow near the opening. The two regimes have very different plasma properties, notably the electron energy distribution function (EEDF) thus allow for device flexibility and control that is very important for technological applications. Increasing the gas pressure leads to elimination of the first regime for gas pressure above few Torr, which confirms that nonlocality of EEDF is responsible for formation of the first regime. This is because the disappearance of bright ball-shaped glow can be explained by the fact that the EEDF in the plasma at higher pressures becoming local, that is, determined by local values of electric field, as opposed to at lower pressure, where an electron can travel without significant energy loss over the discharge gap.

In summary, controlling plasma properties in a dc discharge plasma device with two anodes and a thermionic cathode has been studied in this work. This work was supported by the DOE OFES (Contract No. DE-SC0001939).

References
A wide range of oscillation modes exists in the discharge plasma of a Hall effect thruster (HET). Recent experiments of a HET have shown that the discharge oscillation is associated with the plasma oscillation inside the discharge channel. A hybrid-direct kinetic (DK) simulation is used to model such discharge plasma\(^1\). DK simulation solves the kinetic equations to obtain the velocity distribution functions (VDFs) directly on a discretized phase space. In comparison to the existing particle methods, there are two advantages of using the DK simulation: (1) time averaging technique is not used since there is no statistical noise that arises due to the use of macro-particles; and (2) ionization events are captured every time step on every phase space bins whereas the particle methods cannot capture the ionization in detail due to the memory restriction.

The mode transition of the discharge oscillation in HETs between the global and local ionization oscillation has been studied using the 1D hybrid-DK simulation and experiments\(^3\). The predicted mode transition agrees with experiments in terms of the mean discharge current, the amplitude of the current oscillation, and the breathing mode frequency as shown in Fig. 1.

The stabilization of the global oscillation mode is associated with reduced electron transport that suppresses the ionization process. The balance between the Joule heating and the other energy loss mechanism stabilizes the global plasma oscillation and hence the discharge current oscillation. In the present model, the transport of electronically excited atoms is also taken into consideration. It is indicated that there is a strong correlation between the emitted light intensity and the discharge current.

**References**

Adiabatic Plasma Expansion in a Magnetic Nozzle

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In recent years there has been significant interest in helicon discharges in expanding magnetic fields for applications such as spacecraft propulsion and plasma processing, as well as for studies in basic plasma science. A converging/diverging magnetic field geometry functions like a physical de Laval nozzle for chemical rockets: accelerating the plasma to sonic speeds at the throat and then supersonic speeds in the diverging section of the nozzle. We propose a fluid model for ambipolar ion acceleration in the nozzle that preserves the adiabaticity of the plasma. This adiabatic theory predicts that the change in average electron energy equals the change in plasma potential, providing an important design metric for electric propulsion devices which accelerate ions through magnetic nozzles.

The fluid theory predictions were compared to measurements made in the VASIMR VX-200 experiment which has conditions conducive to ambipolar ion acceleration.\cite{1} A planar Langmuir probe was used to measure the plasma potential, electron density, and electron temperature for a range of mass flow rates (50 – 140 mg / s) and power levels (12 – 30 kW). The linear relationship between electron temperature and plasma potential was observed as predicted (see Fig. 1).\cite{2} The adiabatic theory explained other measurements better than previous, non-local theories.\cite{3}

The adiabatic theory relies on collisionality to rethermalize the electrons and establish a temperature gradient. Coulomb collisions cannot account for the high collisionality, but an ion acoustic instability can enhance the collision frequency.\cite{4} Initial calculations indicate that instability-enhanced collisions can play an important role in the dynamics of plasma in a magnetic nozzle.

\textbf{References}

In this presentation, a survey of collaborative studies that have been facilitated or enabled through the DOE-PLSC will be described. While each study had independent objectives and utilized very different plasma operating regimes, we emphasize common themes that unite the various studies. Specifically, we emphasize the development of diagnostics that are employed for interrogation and characterizing of temporally varying and structurally rich plasma systems to assess the role electron kinetics play these discharges.

Central to the studies described is a common plasma generating setup that provides a flexible platform for generation and interrogation. A rather simple discharge system consisting of a biased electrode and grounded electrode is utilized to generate plasma over a broad range of current densities and pressures will be described. Diagnostics such as Langmuir probe, optical emission spectroscopy, microwave resonance cavity, laser absorption, laser induced fluorescence and laser-collision induced fluorescence are utilized to characterize the spatial and temporal evolution of the pulsed plasma system.

In one study, measurements of optical emission emanating from the argon $3p_5$ (419.8 nm) and the argon $3p_9$ (420.1 nm) are utilized to assess the electron densities, metastable densities and effective values of $E/N$ during the initiation of the plasma with the use of a simplified collisional-radiative model [1]. Measurements of discharge parameters such as current density, electron density and metastable densities are used to test the predictive models derived by collaborators in the PSC.

Other examples of collaboration-driven research centering quantification of helium metastables generated during the launching of a fast ionization wave [2] and the use of the setup to calibrate other laser based diagnostics will be discussed. Finally, an overview of future research efforts will be outlined.

References

Synergetic Effects of Plasma and Material Processes on Self-Organization of Atmospheric Pressure Carbon Arc Discharge

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The atmospheric pressure carbon arc in inert gases such as helium is an important method for the production of nanomaterials [1]. Typical nanosynthesis arcs operate in a dc mode between a graphite anode, which is consumed, and a cathode which can be made from either graphite or a lower melting point material [2]. In spite of many studies, the basic physical processes in this discharge such as cathode electron emission, evaporation and deposition of the anode material, particle and heat transport, and arc instabilities are still not well understood. A lack of understanding of these fundamental processes limits predictive capabilities of existing arc models, including their application for modeling the nanosynthesis. Our current research involves integrated experimental and modeling efforts aimed to develop understanding of the plasma processes and their synergy with material synthesis processes. In addition to arc experiments, we develop in situ diagnostic of nanosynthesis processes [3].

In recent arc experiments, measurements of evaporation and deposition rates, temperature of the electrodes, voltage drop in the arc and the arc current, and ex-situ material characterization of the deposit revealed self-organization of plasma and material processes in the arc discharge [4,5]. In particular, during the arc operation, a carbon deposit is formed on the cathode surface. Electrons emitted from this deposit heat the graphite anode, which evaporates. The carbon ions and atoms travel to the cathode and condense to form the deposit, which is at the high temperature (> 3000 K, Fig. 1) necessary for thermionic emission to support the arc current at the cathode by thermionic electron emission [5].

Our results suggest that for the same operating conditions (gas, pressure, current), the arc can operate in two different regimes of evaporation and deposition of the anode material. The transition between these regimes is determined by the anode diameter (in our experiments ~ 0.8cm). For larger anodes, the evaporation and deposition rates are relatively small and independent on the anode diameter. For smaller anodes, both evaporation and deposition increase dramatically as the anode diameter reduces. This regime appears to be favorable for high yield nanosynthesis. It was suggested that the transition to this regime is due to the formation of the positive anode sheath leading to enhanced power deposition on the anode [6]. This regime is also characterized by the enhanced contribution from the latent heat to the cathode energy balance [4]. Future studies will include detail plasma measurements and numerical simulations of these arc regimes and self-organization which can be important for controlling of nanosynthesis material processes.

References
Atmospheric-pressure plasma jets formed by dielectric barrier discharges and injected into ambient air are repetitively pulsed and so are composed of a sequence of ionization waves (IW). [1] The length of the plasma plume from such devices can be many to 10 cm. The final length is determined by mixing of air with the He plume. With typical applied voltages of a few kV, the mean electric field in the plume can be less than 1 kV/cm which is less than the avalanche field for He. As a result, a conventional -IW (the bullet) cannot be sustained for the observed distances in pristine gas. With repetition rates of a few to tens of kHz, each new plasma bullet propagates in a gas excited and preionized by the previous plasma bullet. In this poster, we discuss results from a computational investigation of repetitively pulsed microplasma jet (1 atm He/O_2 = 99.8/0.2 flowed into air) from the first pulse into an non-ionized gas to a well developed plasma plume. The investigation was conducted with the modeling platform nonPDPSIM.

We found that with each subsequent pulse, the plasma bullet propagates for a longer distance as shown in Fig. 1. In the downstream regions where the mole of fraction of air in the plume is high, most of the electrons attach to O_2 forming O_2^- and O^- . On the next pulse and during propagation of the next bullet a considerable number of electrons are detached from the negative ions in the region of strong electric field in front of the bullet head. With each subsequent pulse, the IW leaves behind a more intensely preionized channel extending for a longer distance. This process continues until the extension of the plume is limited by mixing of air into the He channel.

References
Lipopolysaccharide (LPS), the main component of the outer membrane of Gram-negative bacteria, is notoriously difficult to remove from surfaces by traditional methods. Low pressure studies demonstrating strong radical-induced modifications to LPS motivated studies with an atmospheric pressure plasma jet (APPJ) where neutral species dominate [1]. By adding small N2/O2 admixtures to Ar, we find that the O2 admixture in the APPJ is a major determining factor for both deactivation and surface modification as measured by an enzyme-linked immunosorbent assay and x-ray photoelectron spectroscopy, respectively. N2 admixture without O2 causes minimal deactivation, while N2/O2 admixtures deactivate more with increasing O2 content. For identical O2 feed gas flows, less deactivation occurs when N2 is also added, which demonstrates that nitrogen-based species quench reactive oxygen species (ROS).

As N2 is abundant in the environment, we seek to understand its role in biodeactivation and surface modifications by regulating the interaction of the plasma with the environment. The APPJ is mounted inside a vacuum chamber that can be evacuated and refilled with any gas chemistry. By changing the APPJ geometry, the plasma plume can be either exposed or protected from the ambient. With the plume exposed, increasing concentrations of ambient N2 in Ar reduce surface modifications. With the plume protected, varying ambient N2 concentrations hardly impact the surface chemistry, which shows stronger changes than the exposed case for an identical ambient even though the plasma is farther from the surface when confined. This suggests that the ambient N2 needs to be excited by the plasma to quench ROS. Optical emission spectroscopy and ozone detection confirm that atomic O and O3 are quenched by N2 addition to the plasma. In our collaborative work at Ohio State University, we observed formation of N2* ~1 cm downstream from the nozzle.

After plasma treatments, a new chemical species that we tentatively attribute to NO3, was detected on LPS surfaces. This species has been seen on various carbon-based films exposed under comparable conditions to the APPJ effluents. To determine if this species results from N in the atmosphere, plasma treatments were performed on 193 nm photoresist, which only contains carbon and oxygen. NO3 forms on this surface even if N2 is absent in the feed gas, but interaction of oxygen-based species with N2 in the environment takes place, indicating that plasma-environment interactions create this moiety. APPJ treatments are also compared with a corona discharge to examine the role of ozone and NOx species. An atomistic model describing NO3 formation will also be presented. The authors gratefully acknowledge financial support by the US Department of Energy (DE-SC0005105 and DE-SC0001939) and National Science Foundation (PHY-1004256).

References
Atmospheric pressure plasma jets (APPJ) have been shown to modify surfaces, leading to a variety of potential industrial and medical applications. In this study, we characterized both closely-coupled and remote APPJ treatments of a PMMA-based 193 nm photoresist polymer (PR193) using in situ ellipsometry to monitor film thickness and refractive index in real time. The kilohertz-driven, two-ring electrode APPJ described previously was fed with low admixtures of O₂ and N₂ to Ar [1]. Voltage and current waveforms were collected to characterize electrically the APPJ and measure power dissipation. In addition, high speed photography of the APPJ was conducted in collaboration with Princeton Plasma Physics Laboratory in order to characterize plasma discharge coupling with different controlled environments and the plasma interaction with PR193.

Ellipsometry shows that PR193 etch rates depend on the feed gas chemistry for remote plasma processing. Etch rates are reduced for Ar/O₂ compared with pure Ar and Ar/N₂, which is correlated with a decrease in plasma density. This effect is seen in Figure 1 by comparing a cyclical exposure of Ar and Ar/1%O₂ with Ar. When the plasma is brought closer to the sample surface, the discharge couples with the surface and arcing to the film occurs. This interaction greatly increases the etch rate and introduces major damage to the polymer, which can be observed by the naked eye.

From electrical data and high speed photography we see that the pure Ar discharge exhibits filamentary behavior that is enhanced by O₂ addition and rendered more diffuse by N₂ addition. We also find that dissipated power and breakdown voltage generally increase for admixtures of gases compared to Ar. High speed photography shows that the coupling of the plasma and the environment increases when the environment matches the feed gas chemistry, which causes the plume to extend farther than in open air. While the Ar plume is confined to a single plasma channel, Ar/N₂ admixture branches out into many smaller discharges similar to a Lichtenberg figure. We are also able to spatially correlate damage seen on surfaces with arcing observed using high speed photography. Electrical data in conjunction with high speed photography show that the discharge of the plume into the environment occurs only once per full voltage cycle, despite seeing multiple current peaks in both half cycles of the discharge. This study will be continued by investigating the effect of small H₂O admixtures on plasma characteristics and surface interactions.

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References
Laser-induced Incandescence Diagnostic for In Situ Monitoring of Nanoparticle Synthesis in an Atmospheric Plasma

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A DC arc discharge with a consumed graphite, anode electrode is commonly used for synthesis of carbon nanoparticles, including buckyballs, nanofibers, and nanotubes [1-3]. The graphite electrode is vaporized, leading to nanoparticle synthesis in a low temperature (0.1 – 1 eV), atmospheric pressure plasma. The formation of nanoparticles in this plasma is poorly understood. For example, it is not clear where nanoparticles nucleate and grow in the arc discharge. To tackle this problem, a laser-induced incandescence (LII) diagnostic for in situ monitoring of the nanoparticles’ spatial distribution in the plasma is currently being constructed. The LII diagnostic involves heating the particles with a short-pulsed laser, and measuring the resulting spatial and temporal incandescence profiles on longer timescales [4]. By appropriately modeling the spatiotemporal incandescence profiles, one can measure the particle diameters and volume fraction. LII diagnostics have been extensively used to study soot particles in various backgrounds, including laboratory flames, smokestacks, and engines. However, LII has only recently been applied to study engineered nanoparticles, and has never been applied in a strongly coupled plasma background, such as a carbon arc discharge. Even though the spatial scale-lengths for soot and nanoparticles are similar (10-100 nm), great care is needed in developing an LII diagnostic for monitoring nanoparticles in an atmospheric pressure plasma. Therefore, we will initially calibrate our LII diagnostic by measuring spatiotemporal incandescence profiles of known, research grade carbon particles, including soot and nanoparticles. Preliminary results of this study will be discussed.

References
Long-term Interactions of Atmospheric Pressure Dielectric Barrier Discharge with Wet Tissues

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The interaction of plasmas with liquids is increasingly important in biomedical applications, from sterilization to wound healing. The dose, the time over which plasma is applied, is critical to these applications. For bacterial sterilization, a few seconds to a few minutes of plasma exposure are necessary to achieve the desired effects.[1] Exposure to high doses of plasma can cause cell death.[1] Tissues treated by plasmas are often covered by a thin layer of a water-like liquid. The liquid serves as a filter, processing radicals prior to their reaching the tissue.

In this paper, we report on a computational investigation of the interaction of DBDs with a thin liquid layer covering tissue. These simulations were performed using nonPDPSIM, a 2-dimensional model in which Poisson’s equation, electron temperature equation and transport equations for charged and neutral species are solved. The liquid layer is treated identically to the gas as a partially ionized substance. Liquid evaporates into the gas with a source given by its saturated vapor pressure. The rate of transport of gas phase species into the liquid is determined by Henry’s Law. The liquid layer, typically hundreds of microns thick, is water containing dissolved O\textsubscript{2}aq (aq means aqueous species) and alkane-like hydrocarbons (RHaq).

In the simulations, up to 100 plasma pulses are modeled, including their interaction with the water and transport through the water layer. The DBD is operated with -15 kV applied voltage at 100 Hz for 1 s followed by several minutes of afterglow during which species evolve in the water. Gas phase reactive oxygen and nitrogen species (RONS) solvate when reaching the liquid. Without RH\textsubscript{aq}, NO\textsubscript{3}aq, O\textsubscript{2}aq and hydronium (H\textsubscript{3}O\textsuperscript{+}aq) dominate the water ions with H\textsubscript{3}O\textsuperscript{+}aq determining the pH (shown in Fig. 1). The dominant RONS in the liquid are O\textsubscript{3}aq, H\textsubscript{2}O\textsubscript{2}aq, and ONOO\textsuperscript{-}aq. Dissolved O\textsubscript{2}aq assists with the production of HNO\textsubscript{3}aq and HOONO\textsubscript{aq} in the afterglow. With RH\textsubscript{aq}, ROS are largely consumed at the early stage of plasma treatment, leaving R\textsubscript{aq}• (alkane radical) to reach the tissue. As the fluence of ROS exceeds the area density of RH\textsubscript{aq}, it is possible to completely react the RH\textsubscript{aq} to R\textsubscript{aq}•. At this point, the character of ROS (and R\textsubscript{aq}•) fluxes reaching the underlying tissue changes. This time dependent change in fluences of reactants to the tissue may be important in establishing protocols.

References
Diagnostics for Fundamental Plasma Processes of Chemically Reactive Plasma Interacting with Surfaces with Emphasis on Near-atmospheric Systems*

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The new thrust/focus of the DOE LTPSC in the upcoming fiscal year focuses on understanding laboratory plasma including its chemically reactive species the higher pressures approaching atmosphere. Higher pressure discharges such as atmospheric pressure plasma jets and atmospheric dielectric barrier discharges have been developed to study these plasmas. Naturally, the diagnostics needed to understand these plasmas must be compatible with this high pressure environment. Development of probe theory [1,2] and optical techniques [3] applicable to these dense (near-atmospheric) pressure plasmas is progressing. In this work, we aim to validate the use of our low pressure diagnostics at higher pressures and we identify possible alternatives.

Diagnostics used during collaborations at UMd included a Langmuir probe to determine electron temperature, electron density, and EEDF accurately in the 10mtorr range. Optical emission profiles were measured using a 0.5m spectrometer with a 1200 lines/mm diffraction grating and a PMT. The 420.1nm/419.8nm line ratio yielded a qualitative measure of metastable density in the plasma. We hope to exploit these diagnostics to obtain well-calibrated results at higher pressures.

Diagnostics used during collaborations at SNL include a microwave resonant cavity (MRC) to obtain electron density and temperature, and laser absorption spectroscopy (LAS) to obtain metastable density accurately in the 1torr range. The reduced electric field inside the plasma column was inferred using the plasma current and the results of Pack et. al [4]. The Ar 420.1nm and 419.8nm spectral line intensities were measured using a 0.5m Jobin Yvon Spectrometer, with a 2400 lines/mm diffraction grating and an Andor iStar ICCD camera. These measurements will be interpreted using the extended corona model for argon plasmas to develop diagnostics at higher pressures.

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References
Hall thrusters typically exhibit two principle modes of low frequency oscillations of the discharge, including a longitudinal breathing mode and an $E \times B$ rotational mode (the so-called rotating spoke) [1, 2]. The breathing mode is a distinctive $m = 0$ oscillation of the discharge current ($\sim 10$ kHz), which is usually associated with the ionization instability [2]. Large amplitude breathing oscillations are predicted to affect the plasma plume divergence and thruster efficiency. In this work, we study dynamic perturbations of the ion velocity distribution function induced by breathing oscillations. For that purpose, a novel approach to time-resolving laser-induced fluorescence (LIF) [2] has been developed. The heterodyne technique decomposes the IVDF $f(v) = f^0 + f^1 + \ldots$, where $f^0$ is the time-average of the distribution and the higher-order terms reconstruct the perturbation [3].

In order to validate the heterodyne approach, we operated a cylindrical Hall thruster (CHT) with a modulated applied voltage to induce coherent breathing oscillations. In experiments, the thruster was operated with xenon gas and a dc voltage of 200 V. The amplitude of the modulation signal was varied from 10 to 50 V. An applied sinusoidal signal of 10 V was sufficient to generate coherent discharge oscillations at a frequency of 11 kHz. An LIF signal was collected over all velocity classes of the discharge. The data in Figure 1 display (a) the time-average LIF signal, and (b) the magnitude of the first harmonic component to the perturbation for the discharge. Information relating the phases between fluctuations of ion population levels at each velocity class is also collected, and may be used to reconstruct the full time-dependent behavior of the oscillations. Second and higher-order harmonics are also resolved, and amplitude adjustments of the applied voltage modulation show changes in the levels of ion response. The results demonstrate the successful application of the new time-resolving LIF technique for characterization of breathing oscillations.

**References**

1D Nanodusty Pulsed Plasma Model for the Study and Control of Particle Generation and Growth

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Reactive plasmas present themselves as a unique opportunity for the synthesis of nanomaterials and a huge effort has been devoted to the interpretation of the physical and chemical understanding of aerosol nanoparticle evolution in such plasmas. Indeed, for a well characterized spatially homogeneous aerosol plasma, particles need only be characterized by charge, size and chemical composition. Such reactive plasmas, termed “nanodusty” for the suspension of nonnegligible concentrations of charged nanoclusters, would have an even greater manufacturing advantage over other systems if the formation, growth and transport of such clusters could be controlled. We make use of a 1D plasma model to study such processes.

Our numerical model simulates an RF capacitive argon plasma held within two radially infinite parallel plates. A flow of Argon between the upper a lower electrodes acts as the carrier gas and silane is injected on the upper electrode as a precursor for the particles. The addition of such particles radically changes the equilibrium state of the plasma and at high enough concentrations allows for nucleation of mostly negatively charged clusters and nanoparticles, which remain confined in the plasma as long as the plasma is on and the neutral drag on them from the flow is small.[1]

Spatial profiles of instantaneous electron and positive/negative ion concentrations, electron temperature, plasma potential, and electric field are determined using continuum formulation while nanoparticles are calculated using a Sectional Model that includes Coagulation, Nucleation, Surface Growth and Charging.[2]

It is this combination of continuum and sectional models, each with their own particular time step, that makes this numerical algorithm particularly attractive as it provides a fast and self-consistent reliable tool to study plasmas and their production of particles where the computational time frame to grow particles from Angstroms to tens or hundreds of nm in a plasma is in the order of hours.

This also comes with the advantage given by the flexibility of the numerical scheme to expand a fairly wide range of conditions. For example, in this case, we will demonstrate that one can produce different sized nanoparticles by tuning the frequency and duty-cycles of a pulsing plasma. Simultaneously, we will show that the need to provide accurate chemistry is key to providing reliable results when compared with the experimental results.

References

Kinetic Solvers with Adaptive Mesh in Phase Space: Applications to Electron Kinetics in Low Temperature Plasmas

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An Adaptive Mesh in Phase Space (AMPS) methodology has been recently developed to solve kinetic equations (Boltzmann, Vlasov, Fokker-Planck) for rarefied gas dynamics, radiation transport, and charged particle kinetics in plasmas [1]. In our presentation, we will illustrate the benefits of AMPS for studies of electron kinetics in low temperature plasma.

We will discuss numerical solution to the Vlasov equation for electrons in a collisionless RF sheath. An example of adapted mesh and calculated Electron Distribution Function (EDF) is shown in Fig. 1. At the sheath-plasma edge, at $x/L=1$, the EDF is half-Maxwellian (at $v<0$) with a thermal velocity, $v_T$. The electric field is prescribed in the form:

$$E(x,t) = \frac{2U_0}{L} \left( x - s(t) \right),$$

where $s(t)$ is the sheath boundary position, and $U_0$ is the maximal value of the potential drop in the sheath, $U_0 \gg \frac{mv_T^2}{2e}$. Electron dynamics is characterized by the dimensionless frequency $\Omega = \omega L / v_T$, where $\omega$ is the angular frequency of the field oscillations. Fig. 1 shows results for $\Omega = 0.1$, when an electron “wave riding” effect is observed. Due to this effect, at some moments during RF period, electrons gain considerable energy after being repelled by the moving sheath boundary. Using AMPS allows high-resolution of the EDF dynamics with minimum number of computational cells.

We will also discuss effects of elastic and inelastic collisions on the EDF formation in DC and RF discharges under different regimes [2]. Similarities between electron kinetics in capacitively coupled discharges and rarefied gas dynamics near oscillating walls [3] will be outlined. We will analyze peculiarities of the particle kinetics at different values of the dimensionless frequency $\Omega$, and the Knudsen number $Kn = \lambda / L$, where $\lambda$ is the mean free path. Fundamental differences between the gas dynamics and plasma physics phenomena will be clarified.

References
Numerical Simulation of 2D Capacitively Coupled RF Plasma for the Synthesis of Silicon Nanocrystals

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Silicon nanocrystals are of interest due to applications in light emission, photovoltaics, nanoelectronics, and biomedical. Liquid-phase, gas-phase, and plasma techniques can produce SiNCs. Nonthermal plasmas have the advantage over other processes to produce narrower size distribution due to unipolar charging [1]. The Kortshagen group (University of Minnesota) has produced many papers on experimental SiNCs production. This work models their reactor [2] which can tailor nanometer-size SiNCs. Comparison with experiments will be done together with the Kortshagen group. Numerical simulations are conducted through collaboration between the Girshick group (Aerosol-Plasma Model [3]) and the Kushner group (Hybrid Plasma Equipment Model—HPEM) [4].

Experimental results show silicon nanoparticles deposition on tube walls, which implies that nanoparticles are formed over the entire tube, even a few centimeters above the top electrode. Preliminary numerical simulations confirm this observation. Dense plasma occurs from the pair of ring electrodes to the vacuum chamber, and a dimmer plasma above the electrode, as observed in [2]. Nucleation is assumed to occur by a negative ion pathway, \( \text{Si}_2\text{H}_4^- \) and \( \text{Si}_2\text{H}_5^- \). Their densities peak downstream of the electrodes. Radical deposition on nanoparticles leads to surface growth. Radical density also peaks downstream of the electrodes. Nanoparticles grow due to coagulation and surface growth. Figure 2 shows nanoparticle size distribution at 5cm from the outlet from preliminary results (not fully converged). Power supply, flow rates, gas composition are plasma parameters that will need to be studied to see their effects on nanoparticle nucleation and growth.

References
Ignition Delay in a Faraday-shielded Dual Source Pulsed Chlorine Plasma

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Capacitive coupling in inductively coupled plasmas (ICP) is often undesirable because it causes ions to accelerate across sheaths and bombard processing chamber walls at high energies, causing sputtering contamination and heating of the walls. A Faraday shield can be employed to reduce capacitive coupling. However, in the presence of a Faraday shield, it is challenging to operate a pulsed chlorine plasma with a long afterglow duration, as most of the electrons are lost by dissociative attachment in the afterglow, forming an ion-ion plasma.

To operate Faraday-shielded pulsed plasmas in electronegative gases (such as chlorine) with long afterglow duration, a dual plasma source was developed, consisting of a main ICP in tandem with an auxiliary ICP. The two sources are separated by a grounded metal grid. Power to the main ICP is pulsed at a frequency of 1 kHz. The auxiliary ICP is operated in continuous wave mode, providing electrons through the grid that help ignition (start of the active glow) of the main plasma. Langmuir probe and optical emission spectroscopy (OES) were employed to record plasma parameters and optical emission from chlorine atoms at 8375 Å. Ignition delay was observed with duty cycles larger than ~60%, reaching a maximum delay at a duty cycle of ~99%. At this condition, the main ICP ignited with a delay of more than 500 µs after RF power was turned on. The ignition delay fell to zero at a duty cycle >99.7%.

Figure 1(a) shows the charged species densities and electron temperature for a plasma pulsed at 99% duty cycle of lower ICP. After the power is turned off at t= 990 µs, the electron density and temperature drop precipitously. After the power is turned back on at t=0 µs, the electron density continues to decay to a value lower than the electron density prevailing when only the upper ICP is powered (black dashed line), while the electron temperature slowly increases. At ~500 µs, plasma ignition occurs.

It was found that the ignition delay time decreases with increasing auxiliary ICP power or with decreasing main ICP power (Figure 1(b)). The ignition delay may be attributed to low electron density in the main ICP at the end of the afterglow, causing poor power transfer when the RF power is turned back on, preventing the start of the active glow. The flux of seed electrons from the auxiliary ICP restores the electron density, thereby improving the power transfer in the main ICP and allowing plasma ignition. Similar results were also observed using other electronegative gases such as SF₆ and CF₄/O₂ mixtures.
Fluorocarbon Assisted Atomic Layer Etching of SiO$_2$ and Selectivity over Si Using Cyclic Ar/C$_4$F$_8$ Plasma

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There is great interest in establishing directional etching methods capable of atomic scale resolution for fabrication of highly scaled electronic devices. Recently, controlled etching of SiO$_2$ at the Angstrom-level based on steady-state Ar plasma, periodic injection of a defined number of C$_4$F$_8$ molecules, and synchronized plasma-based Ar$^+$ ion bombardment has been shown [1]. Controlled etching is based on deposition of a thin (≈ several Å) reactive fluorocarbon (FC) layer on SiO$_2$ enabled by precise C$_4$F$_8$ injection. For low energy Ar$^+$ ion bombardment conditions, the physical sputter rate of SiO$_2$ vanishes, whereas SiO$_2$ can be etched when FC reactants are present at the surface. Plasma conditions have been characterized during cyclic exposure using a Langmuir probe. EEPFs at various characteristic times during one cycle and for continuous addition of 2% C$_4$F$_8$ into Ar plasma can be seen in Fig. 1. Continuous precursor addition has a higher C$_4$F$_8$ concentration than periodic injection. Plasma potential, electron temperature, and electron density have been extracted and compared. The C$_4$F$_8$ injection has a short, significant impact on the plasma properties within each cycle and a small impact for longer time scales, i.e. from cycle to cycle. Observed trends in plasma properties agree with continuous precursor addition. Additionally, this cyclic approach was used to investigate the transition from SiO$_2$ to Si etching employing SiO$_2$-Si-SiO$_2$ layers. Si etching and the selectivity of SiO$_2$ over Si is investigated as a function of FC surface coverage, ion energy (20 to 30 eV), and etch step length using in situ ellipsometry. Time-dependent etch and deposition rates are compared for Si and SiO$_2$.

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References:
Time-Resolved Correlation of Excitation Processes of a Pulsed Argon Discharge Column at 1% Atmosphere*

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Direct measurement of the intensities of the Ar 420.1nm and 419.8nm spectral lines, independent measurement of metastable-atom density, electron density, and electron temperature, inferred measurement of reduced electric field, and assumptions of the shape of the electron energy distribution in a 1-torr argon discharge are correlated within the extended-corona-model framework applied to the late stage of a pulsed discharge-tube plasma to determine the reaction rate constants $k_{ij}$ that are used to predict the 420.1nm to 419.8nm line ratio [1]. The model predicts post-transient metastable density. The diagnostic use of optical emission spectroscopy (OES) to follow experimental conditions from start to end of the transient phase of the pulsed discharge is discussed.

The Ar 420.1nm and 419.8nm spectral line intensities were measured using a 0.5m Jobin Yvon Spectrometer, with a 2400 lines/mm diffraction grating and an Andor iStar ICCD camera. The density of 1s5 argon metastable atoms (in Paschen’s notation) in the discharge was measured by laser diode absorption spectroscopy using a tunable diode laser. Electron density and electron temperature measurements were obtained using microwave techniques [2]. Plasma current was measured and combined with the results of Pack, et al. [3] to infer the local value of reduced electric field ($E/n$). The value of reduced electric field and the results of Adams et al. [1] were combined to infer the values of the reaction rate constants.

Insight into hard-to-measure or expensive-to-measure plasma conditions and their time dependence is afforded by invoking plausible assumptions regarding the shape of the electron energy distribution function (EEDF) which evolves and may or may not be consistent with assumptions at all times. The next step is to validate assumptions regarding the EEDF shape in both the transient and post-transient discharge.

References

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Photo-assisted Etching of Silicon in Halogen-containing Plasmas

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With an increasing need to reduce the feature size to <14 nm, highly precise techniques that can etch semiconductor materials without causing damage are required. In a recent study, it was found that etching of silicon occurs even below the ion assisted etching energy threshold. This sub-threshold etching was attributed to photo assisted etching dominated by VUV photons generated in the plasma[1].

Cl₂, Br₂, HBr, and HBr/Cl₂ feed gases diluted in Ar were used to study photo assisted etching of p-type Si(100) in a RF inductively coupled, Faraday-shielded plasma. Etching rates were measured as a function of ion energy. Etching at ion energies below the threshold for ion-assisted etching was observed in all cases, with Br₂/Ar and HBr/Cl₂/Ar plasmas having the lowest and highest sub-threshold etching rates, respectively. Sub-threshold etching rates scaled with the product of surface halogen coverage (measured by XPS) and Ar emission intensity (7504Å). Etching rates measured under MgF₂, quartz, and opaque windows, or biased grids, showed that sub-threshold etching is due to photon-stimulated processes on the surface, with VUV photons being much more effective than longer wavelengths.

Characterization of etched surfaces using Scanning Electron and Atomic Force Microscopy (SEM and AFM) revealed that photo-etched surfaces were very rough, quite likely due to the inability of the photo-assisted process to remove contaminants from the surface. Photo-assisted etching in Cl₂/Ar plasmas resulted in the formation of 4-sided pyramidal features with bases that formed an angle of 45° with respect to <110> cleavage planes, suggesting that the photo-assisted etching process is sensitive to crystal orientation, at least for chlorine. Possible synergistic mechanisms of plasma-photoassisted etching involving the formation of energetic electron-hole pairs, band bending and hole-catalyzed reactions will be discussed.

Reference
Controlling VUV Photon Fluxes in Pulsed Inductively Coupled Plasmas: Rare Gas Mixtures

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Low pressure (a few to hundreds mTorr) inductively coupled plasmas (ICPs), as typically used in microelectronics fabrication, can produce UV/VUV photon fluxes to the substrate comparable to or exceeding the ion fluxes.[1] These VUV photon fluxes are desirable in applications such as sterilization of medical equipment and are unwanted in many materials fabrication processes due to damage to the devices.[2] Under select conditions, VUV fluxes may stimulate etching or synergistically combine with ion fluxes to modify polymeric materials.[3] In this regard, it is desirable to control the magnitude of VUV fluxes, the ratio of VUV fluxes to that of other reactive species, such as ions, or discretely control the VUV spectrum.

In this work, we discuss results for VUV fluxes produced low pressure ICPs sustained in rare gas mixtures through a computational investigation. Simulation were performed using Hybrid Plasma Equipment Model, a 2-dimensional plasma hydrodynamics model which addresses radiation transport using a Monte Carlo Simulation. The control of VUV fluxes is investigated through pressure, gas mixture and the use of pulsed power.

We found out that photon fluxes can be controlled with the duty cycle of the pulsed power, which is consistent with previous work in halogen-containing plasmas. For example, the time-averaged photon fluxes produced in argon plasmas are sensitive to pressure, reaching a peak value between 10 to 15 mTorr. The spectra of the fluxes are discretely tunable with the composition of the gas mixture. In Penning mixtures, such as He/Xe, where the resonance photon energy of the buffer gas exceeds the ionization potential of the heavier rare gas, the higher energy resonance photons are largely consumed by photoionization.

References
Non-Stationary Charging Dynamics of an Inhomogeneous Granule-Plasma Multi-phase System

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Gyro-phase drift is a sensitive indicator of dust grain charging rate [1]. The trajectory of a dust grain, radius $a$, can be modelled semi-analytically for an abrupt inhomogeneity [2]. For an abrupt inhomogeneity, two neighboring regions are characterized by two respective sets of plasma parameters and corresponding in-situ equilibrium charge states. The grain charges or discharges with each gyro-excursion between regions at a characteristic charging time $\tau_{ch}$. We assess gyro-phase drift and direction for the Orbit Motion Limited (OML), Patacchini-Hutchinson electron current [3], and Gatti-Kortshagen ion current [4] charging models for a given set of plasma parameters. Of particular interest is the regime where Debye length $\lambda_d \sim a$, $\lambda_i \sim \lambda_d$, and $a/R_{le} \sim 1$, because under these conditions, the effects of collisionally-enhanced ion current from ion-neutral charge exchange collisions, electron magnetization effects in the grain sheath, and finite shielding length are important. Here, $\lambda_i$ is ion mean-free-path and $R_{le}$ is gyro-radius. The three models yield different gyro-phase drift magnitudes, demonstrating from a theoretical standpoint that gyro-phase drift discriminates among different charge models. Neutral drag force, or Epstein drag, is included in our analysis, and it is assumed that the perpendicular dust grain velocity is small with respect to the thermal speed of neutrals. The application of these theoretical results to dust confinement and model validation in the Auburn Magnetized Dusty Plasma Experiment [5] is assessed through simulations.

References


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Control of Ion Energy Distribution with Phase Shifting in Multi-frequency Capacitively Coupled Plasmas*

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Anisotropic etching for microelectronics fabrication is accomplished by energetic ion bombardment in chemically enhanced sputtering. One challenge is being able to control the ion energy-angular distributions (IEADs) onto the surface of the wafer to selectively activate desired processes, which is advantageous for maintaining the critical dimension (CD) of features. Capacitive coupled plasmas (CCPs) powered by non-sinusoidal waveforms and or using multiple frequencies are strategies employed to provide flexible control of IEADs which produce high selectivity and uniformity. Varying relative voltages, powers and phases between multiple frequencies that differ by integer multiples have demonstrated potential control mechanisms for the IEADs and optimization of etching profiles.

In this paper, we report on computational and experimental investigations of IEAD control in multi-frequency CCPs where the phase between the frequencies is used as a control variable. The rf frequency and its harmonic frequencies are both applied to the wafer substrate. A third frequency may be applied to the top electrode for additional control of the plasma density. The Hybrid Plasma Equipment Model (HPEM) was employed to predict plasma properties and IEADs. The operating conditions are 20 mTorr, with dual CCP frequencies of 15/30 MHz and triple frequencies of 15/30/60 MHz.

We find that by changing the phase between the applied rf frequency and its second harmonic, the Electrical Asymmetric Effects (EAE) is significant and can shift the dc self-bias as shown in Fig. 1. When phases are selected to be symmetric around 90 degrees (that is, \(90 \pm \Delta \phi\)), the EAE effect produces about the same dc bias. However, the shape of the IED is a sensitive function of the precise value of the phase shift. For example, there are systematic exchanges of peaks of the IED depending on whether the phase is +\(\Delta \phi\) or -\(\Delta \phi\) (both having the same dc bias). Similar trends are observed experimentally.

References
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