White Paper for *Frontiers of Plasma Science Panel*

Date of Submission:	***June 19, 2015***

Indicate the primary area this white paper addresses by placing "P" in right column.
Indicate secondary area or areas by placing "S" in right column

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

Indicate type of presentation desired at Town Hall Meeting.

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

Title:	The Physics of Nanodusty Plasmas
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(Limit text to 3-pages including this form. Font Times Roman size 11.
1 page of references and 1 page of figures may also be included. *Submit in PDF format.*)

- ***Describe the research frontier* and importance of the scientific challenge.**

Nanodusty plasmas contain clouds of nanoparticles whose presence alters the plasma properties in a substantial manner. They first attracted attention[1] when homogeneous nucleation was discovered to form nanoparticles in semiconductor processing plasmas. As these nanoparticles presented a contamination problem, they were called "dust" and research aimed at avoiding them. Such a viewpoint is still timely in many areas of plasma processing as well as in fusion research, where particulate formation was associated with materials sputtered from reactor walls.[2] While important applications motivate the use of nanodusty plasmas, their physics poses a number of basic science challenges. Nanodusty plasmas likely exceed in

their complexity many of the traditional areas of plasma research. In the following, we discuss some of the most important challenges and research needs.

There are fundamental research needs in understanding the dynamics of nanodusty plasmas at both the microscopic and macroscopic level. One example of the poorly understood microscopic dynamics is the unusually high kinetic temperature of nanoparticles that is observed in experiments.[3,4,5] One may speculate that Coulomb interactions with charged species are the source of this kinetic heating.[6] Coulomb collisions with drifting ions at sheath boundaries may be particularly effective in kinetically heating the nanoparticles. Both the mechanisms of kinetic heating of individual nanoparticles in plasmas as well as the "heat conduction" within the nanoparticle fluid require further research.

At a more macroscopic view, nanodusty plasmas are highly complex "multi-fluid media". Plasma problems often deal with a relatively limited number of species: electrons, ions, and neutral species. The level of complexity increases in chemically reactive plasmas, which often contain tens of important species and require the consideration of hundreds of reactions. Nanodusty plasmas add further levels of complexity, as they usually comprise a wide spectrum of nanoparticle size and charge states. To model such systems, 1-nm particles need to be treated differently than 10-nm particles, and particles carrying 1 elementary charge behave differently than particles having 10 charges. Modeling techniques for such systems are just in their infancy and further research is essential to advance the predictive capabilities to nanodusty plasma models.[7,8] Furthermore, the problem of chemical nucleation of nanoparticles[9] through either ion-induced or neutral reactions remains essentially unsolved.

- ***Describe the approach to advancing the frontier and indicate if new research tools or capabilities are required.***

Plasma-nanoparticle surface interactions: Nanodusty plasmas are complex multi-phase systems. Interactions of plasmas with surfaces in the solid or liquid phase are arguably among the most complex problems of plasma physics. In many systems, plasma-surface interactions are limited to the reactor walls and constitute mere boundary conditions for the bulk plasma. Such a separation is not possible in nanodusty plasmas, as the solid nanoparticle phase is interspersed within the bulk plasma, and interactions of the plasma with nanoparticle surfaces affect the plasma throughout its volume. The impact of effects such as secondary or photo-electron emission from particle surfaces on the plasma conditions is poorly understood and a fundamental research need. Experimental studies may utilize different nanoparticle materials with different efficiencies for electron emission to explore these effects. Numerical studies can also shed further light on these problems.

Plasma-nanoparticle interactions are also crucial from the viewpoint of the solid nanoparticle phase. Energetic reactions on nanoparticle surfaces cause non-equilibrium to develop between the nanoparticle intrinsic temperature and the neutral gas temperature.[10] The elevated nanoparticle temperatures and the bombardment of nanoparticles with plasma species are believed to play an important role for many physical properties of nanoparticles.[11] These interactions are only vaguely understood and require further research. Atomic scale modeling of nanoparticle interactions with plasma species, for instance through molecular dynamics simulations,[12] may complement experimental studies that relate plasma conditions to microstructural properties of the nanoparticles produced.

Nanodusty plasma dynamics: Research needs exist at both the microscopic and macroscopic level. Understanding the unusually high kinetic temperature of nanoparticles will require both experiments and modeling studies. Experimental studies may rely on laser light scattering, for instance, with techniques similar to dynamic light scattering. Such studies may enable spatially resolved measurements of the nanoparticle kinetic temperature that may give insight both into the heating mechanisms as well as the "heat conduction" in the nanoparticle fluid. Numerical studies, for example with particle-in-cell simulations,[13] may also provide important insights into this problem.

At the macroscopic level, studying the "multi-fluid" character of nanodusty plasmas remains a priority. Experimental studies will rely on well designed experimental systems that enable sophisticated diagnostics of the plasma species, chemical species, and the nanoparticle fluid. Standard plasma diagnostics such as probes often require adaptation to the chemically reactive environment.[14] Studies of the nanoparticle component may rely on *in situ* measurements such as laser scattering or particle mass spectroscopy; *ex situ* measurements may include electron microscopy, or X-ray diffraction. On the modeling side, numerical models that tightly couple self-consistent plasma models with space- and time-resolved chemical nucleation models and aerosol models will have a great impact on elucidating the "multi-fluid" character of nanodusty plasmas.

Bridging the pressure gap: The vast majority of nanodusty plasma studies has focused on low pressure systems with gas pressures often in the range of a few 100 Pa. As nanodusty plasmas attract increased attention for the synthesis of materials, a move to atmospheric pressure becomes desirable for many practical applications. First reports of nanodusty plasmas at atmospheric pressure already indicate that many of the basic mechanisms, which are still poorly understood even at low pressure, are very different at high pressure. For instance, it appears that the intrinsic nanoparticle temperature is, not surprisingly, much more strongly coupled to the neutral species.[15] Hence achieving high intrinsic temperatures may require much higher plasma densities than at low pressure. Both numerical and experimental tools to study nanodusty plasmas at atmospheric pressure are just starting to be developed and require significant further research.

- ***Describe the impact of this research on plasma science and related disciplines and any potential for societal benefit***.

In recent years, researchers realized the intriguing abilities of nanodusty plasmas to serve as sources for useful nanoparticle materials with well controlled properties. The plasma environment offers a number of attractive properties that allow for the generation of nanoparticle materials that are otherwise hard to produce by other means. Among these are the generally high temperatures that nanoparticles can attain within plasmas, enabling the generation of nanocrystals of high melting point materials. The negative nanoparticle charge in plasmas suppresses particle coagulation and enables the synthesis of highly monodisperse nanoparticles. The negative charge also confines nanoparticles in the reactor, very different from neutral gas synthesis, in which particles are often quickly lost to the reactor walls due to diffusion.

These unique capabilities of nanodusy plasmas have already enabled technological advances such as the generation of nanometer-sized diamonds (Fig. 1),[16] record group IV nanocrystal based light emitting devices (Fig. 2),[17] stable silicon-nanocrystal anodes for lithium ion batteries (Fig. 3),[18] and high-mobility transparent conductive oxide films based on zinc oxide nanocrystals (Fig. 4).[19] Furthermore, unique plasma-synthesized nanocrystals have also enabled progress in basic science, such as the recent reformulation of the famous Mott criterion for the metal-insulator transition into a criterion for highly doped films of plasma-produced nanocrystals (Fig. 5).[20]

We expect that studies of nanodusty plasmas will expand our understanding of these highly complex plasma systems and thus make a strong contribution to the progress of plasma physics as a whole. The unique materials that can be produced with plasmas will also impact other fields of science, including materials science and solid state physics. Furthermore, we expect plasma produced nanocrystals to enable significant technological advances in fields including renewable energy generation, conversion, and storage, biotechnology and human medicine.

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Figures (maximum 1 page)

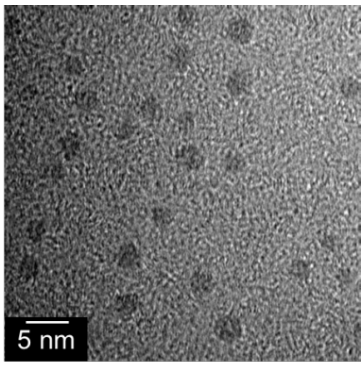

Figure 1: Nanometer-sized diamonds synthesized in an atmospheric pressure microplasma.

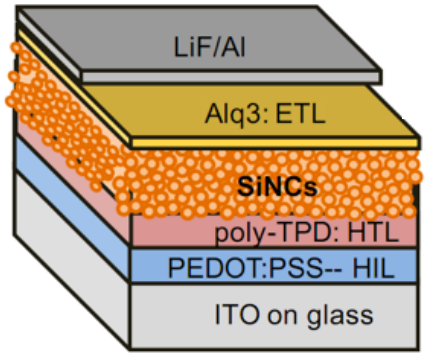

Figure 2: Schematic of a light emitting device based in plasma produced silicon nanocrystals. This device achieved an external quantum efficiency of ~9%.

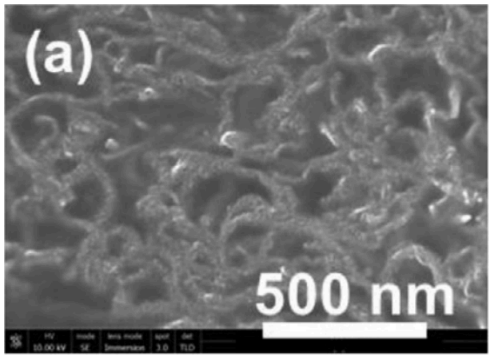

Figure 3: Lithium ion battery anode based on plasma produced silicon nanocrystals.

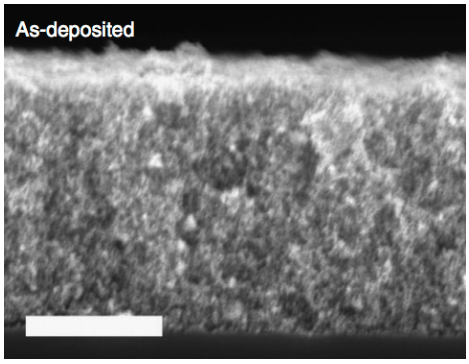

Figure 4: A film of plasma produced zinc oxide nanocrystals. After post-processing, such a film can attain properties that are competitive with commercial ITO films for transparent electrode materials.

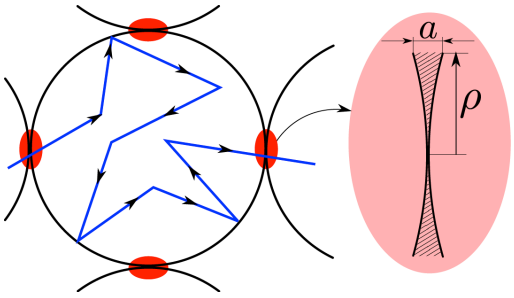

Figure 5: Schematic of diffusive transport in films of degenerately doped (i.e. metallic) silicon nanocrystals. Transport in such granular films shows a metal-to-insulator transition which is significantly different from bulk material and requires a reformulation of the famous Mott criterion.