The Multi-Phase Game: Liquid Wets Gas, Gas Dilutes Plasma, Plasma Surrounds Liquid and all Together They Produce Solid Nanorocks

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The modern low temperature and atmospheric pressure non-equilibrium plasma has made tremendous advances in a relatively short time with respect to providing added value across a broad range of important applications. However, the complexity of the plasma and its interaction with environments and surfaces resists detailed experimental exploration. Distances and timescales for important reactions are often on the nanoscale and measurements are often beyond our meagre resources. A typical plasma-liquid setup has thin interface regions above and below the nominal interface boundary. Apart from charged, atomic and molecular species, these regions also inevitably contain macroscopic objects such as liquid droplets and possibly plasma-filled bubbles. In certain experiments, they can also contain solid nanoscale and microscale particles. Between the macroscopic and the molecular, we also have clusters. This is a truly complex problem in multiphase systems and to date we have tended to minimize the importance of these "extra" phases. Of course, when these objects are large enough, they can be considered as traditional surfaces but on the microscale or lower, these solid/liquid objects can be totally surrounded by plasma, exchanging charge, energy and mass. The plasma-filled bubble is similarly surrounded. We consider the question as to whether it is valuable, or even possible, in this context to study the plasma interaction with individual objects at microscale or lower. In our recent work, we attempt to study such interactions with individual liquid microdroplets, with nanoparticle ensembles and with nanoparticles inside droplets and in this talk, the focus will be on microdroplet studies.

The plasma-microdroplet can act as an individual laboratory experiment enabling a range of conditions that cannot otherwise be achieved. For example, precise control of the gas environment becomes a new parameter while flow and convective forces onto the liquid surface can be minimised or even eliminated. The spherical symmetry enhances the computational capabilities as does the reduced geometric dimensions. The liquid surface is irradiated by ultra-low energy electrons and at these energies the interaction of electrons with liquid is thought to be one of the primary reasons that plasma – liquid interactions are so advantageous, able to locally create high densities of active species that can treat cancer cells, destroy antibiotic resistant microbes, encourage seed germination in agriculture and synthesise novel chemicals and nanomaterials. On the gas side, an evaporation shell layer may contain a cocktail of positively and negatively charged clusters along with a very high localised vapour density. We can speculate that this may lead to amplified non-equilibrium gas-phase chemistry, the products of which may be sequestered by the droplet boundary. By controlling the plasma exposure time of the droplets to the sub-millisecond range, which is not possible with any other steady-state system, we may gain insight into the transport and mass accommodation of radical species before the onset of Henry's Law. The disadvantage is of course the very small droplet volumes and their velocity.

We will report current microdroplet results and highlight new directions via Plasma-XPS where we hope to study the plasma induced chemistry of liquid droplets and solids on a monolayer scale via instantaneous freezing. We also aim to explore optical and tunable infrared absorption (1um – 12um) of droplets and their surrounding vapour layer, in flight.