

69g Interfacial Electro-Kinetic Phenomena Due to Plasma Polarization

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We report a new class of electro-kinetic phenomena that arise when the interfacial double layer is polarized by plasma in the presence of a high-frequency (>1 kHz) gas phase AC electric field. Such polarization can be due to ionization reaction at the tip of a drop electrode, where the field is sufficiently high to effect gas-phase ionization, or due to interaction of the electric field with the bulk plasma generated at a different source. For both polarization mechanisms, the conductivity or permittivity of the liquid is irrelevant as long as they are significantly higher than the gas phase. However, gas-phase ionization reaction to generate plasma, either at the drop electrode or at a different point electrode, is of paramount importance. Gas flow and trace amounts of He and Ar, which are known to catalyze plasma generation, can sensitively affect the resulting electro-kinetic phenomena (D. Lastochkin and H.-C. Chang, "A High-Frequency Electrospray Driven by Gas Volume Charges", *J. App. Phys.*, in press).

The AC spray mechanism is shown to be dominated by longitudinal flow within the drop that is driven by a gradient in the normal Maxwell stress. With a secondary DC field to deflect the ejected drops, the interfacial ionization reaction is experimentally shown to produce a net negative charge at a drop electrode at sufficiently high frequencies. This uniform interfacial net charge minimizes the tangential component of the interfacial field and hence reduces the tangential conduction current that accumulates charges at the tip over each half cycle. The short frequency further diminishes charge accumulation. As a result, the classical Taylor cone for a DC field or a sufficiently low frequency AC field, which has (theoretically) a singular field and infinitely high charge density at the tip, is never observed. Instead, the AC polarization is theoretically shown to favor a cylindrical geometry in the micro-jet that forms when the drop transforms into a spray beyond a critical r.m.s. voltage (L. Y. Yeo, D. Lastochkin, S-C. Wang, and H-C. Chang "A New AC Electrospray Mechanism by Maxwell-Wagner Polarization and Capillary Resonance" *Phys. Rev. Lett.* 2004). The mostly normal interfacial field due to the net plasma charge produces a normal Maxwell stress with a specific scaling with respect to the drop radius. Using a long wave theory, it is then shown numerically as well as analytically that the jet velocity has an inverse relationship with viscosity and the drop size has a linear scaling with respect to the applied field. These scaling results are consistent with our experimental measurements. The drops of this AC spray are neutrally charged within the liquid and hence, unlike their DC counterparts, do not undergo further Rayleigh fission into smaller drops. The stability of the drops allows us to use this AC spray to encapsulate drugs and other bio-materials by spraying polymer solutions (L. Y. Yeo, Z. Gagnon and H.-C. Chang, "AC electrospray biomaterials synthesis", *Biomaterials*, in press).

We also report a novel interfacial flow resulting from the tangential Maxwell stress due to a finite tangential component of the interfacial electric field. Significant tangential electric field and Maxwell stress exist because the interfacial polarization is not due to net accumulation of a local ionization reaction product, as is the case for the AC spray, but is due to AC charging of the bulk plasma generated elsewhere. The interfacial flow driven by this tangential stress is largest at an optimum frequency corresponding to the inverse RC time of the gas phase, when the period is sufficiently large to allow charging of the double layer but not excessively high such that the external field is screened. Assuming a rigid interface, we derive a formulation for the gas electric field sustained by a point electrode and the resultant interfacial gas flow on the flat interface is shown to be a planar vortex pair confined to a thin boundary layer above the interface. By allowing for finite liquid viscosity, interfacial stress transmission produces an image planar vortex in the liquid that penetrates deep into the bulk liquid. We have experimentally demonstrated the existence of this vortex pair. This vortex pair suffers instability at high frequencies and voltages despite the low Reynolds numbers. Instead of the usual inertial vortex breakdown, these instabilities are due to high Peclet number tangential convection in the interfacial double layer, which tends to neutralize the double layer and render its polarization non-uniform. Vortex

oscillation and vortex shedding are observed in a sequence of bifurcations. Nevertheless, within a large voltage and frequency window, a robust vortex is shown to exist and is used for micro-fluidic reagent mixing or particle filtration.

These two new electro-kinetic phenomena due to interfacial polarization by plasma hence differ in the relative dominance of tangential and normal electric field and Maxwell stress, and in the resulting interfacial flows. They exhibit pressure-driven flow, interfacial drag, capillary pinchoff, vortex instability etc as in other viscous flows driven by different body and interfacial forces.