Liquid micro-droplet effects in a plasma

Joshua Holgate. Supervisor: Dr Michael Coppins

Department of Physics, Blackett Laboratory, Imperial College London, London SW7 2AZ

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Abstract

This essay describes the current state of research into micrometre-scale liquid droplets in an ionised gas, usually referred to as a plasma. Such systems are a common occurrence in industrial processes, but have received little interest until recently. Electrostatic breakup is a significant mechanism for disrupting these droplets, but the effect of many plasma parameters on this mechanism has not yet been studied. My PhD project is primarily concerned with developing existing theories of electrostatic breakup of these liquid micro-droplets. I also intend to study the dynamics of micro-droplets in plasma. Some preliminary results for droplet breakup in an electric field are presented here, before discussing key areas for future work.

1 Introduction

When a gas is heated to a sufficiently high temperature then its constituent atoms begin to ionise, forming a new state matter known as a plasma. A plasma is a mixture of electrons, positive ions and neutral atoms whose behaviour is dominated by electromagnetic forces. Plasmas are very common; they exist naturally on Earth, for example in the ionosphere, flames and lightning, and in astrophysical objects such as stars and the interplanetary medium. Indeed it is often claimed that over 99% of the matter in the universe is in the plasma state [1]. Plasmas also have an increasing number of technological uses: in electronic chip manufacturing, welding and cutting tools, fluorescent lighting, televisions, spacecraft propulsion and in controlled fusion energy reactors.

Most plasmas contain impurities in the form of micrometre-scale particles known as dust. Research into dusty plasmas is a relatively new area of applied physics, becoming important in the early 1990s following observations that dust contamination can cause damage to microchips during electronic chip manufacturing [2]. Despite significant progress in dusty plasma research, many unresolved questions remain; for example the theoretical description of large (>100 μm) and non-spherical grains still requires improvement [3]. The theories developed so far have assumed that the dust grains are solid, but it has recently been noted that dust can take the form of liquid droplets in many situations, significantly changing the particle behaviour [4]. Examples include:

- plasmas for microchip manufacturing, in which particles can grow through condensation to the liquid state [5];
- the use of micro-droplets as carriers for plasma-enhanced chemical vapour deposition (PECVD) [6];
- magnetic confinement fusion plasmas, in which the existence of molten metal particles can be inferred from spherical dust collected in many experiments [7];
- plasma spraying, where liquid droplets are injected into a plasma jet for material coating [8];
- the use of alcohol droplets injected into plasma as a diagnostic technique [9].

Given the clear technological importance of liquid droplets in plasma, extension of dusty plasma theory to the liquid state has been identified as a vital area of future research [3]. Accordingly, my PhD project will aim to advance current liquid micro-droplet theories.

In this essay two main differences between liquid and solid particles will be discussed: stability and dynamics. It has been shown that liquid droplets acquire charge from the plasma and are susceptible to electrostatic breakup [4]; this may be desirable to reduce impurities in the plasma, or detrimental in the cases where droplets are intentionally injected into the plasma. In section 2 this theory of droplet breakup will be reviewed and extended to include the effects of an external electric field, and future work to include magnetic field, plasma flow and spinning droplet effects will be discussed. Section 3 will consider how the viscosity and deformation of liquid droplets might lead to dynamical differences when compared with solid particles. Conclusions are drawn in section 4.

2 Droplet stability

2.1 Review of current theory

When a dust grain enters a plasma it is bombarded by electrons and positive ions. As the electron mass is significantly smaller than the ion mass, the electrons are more mobile and have a higher flow rate to the dust, giving it a negative charge. After a short period of time (of order nanoseconds, known as the charging time) the ions become attracted to the negatively charged dust, so ion flow rate increases until it equals that of the electrons. This process charges the dust to a negative potential ϕ_d which, for small, spherical dust grains, is well described by the orbit-motion limited (OML) theory [10] and is independent of the size of the grain. In the case of a liquid droplet, if ϕ_d exceeds a critical value then the droplet is electrostatically disrupted [4].

The problem of electrostatic breakup of a conducting liquid droplet *in vacuo* was originally solved by Lord Rayleigh [11]. An outline of the methods used can be found in [12] and [13]. By considering small perturbations to the droplet surface due the surface tension and electrostatic forces, he found the normal modes of droplet oscillations and obtained a dispersion relation for ω^2. When $\omega^2 < 0$ the droplet oscillations become unstable and the droplet breaks up. The smallest charge for which this happens is given by

$$Q_c = 8\pi(\epsilon_0\gamma a^3)^{1/2} \tag{2.1}$$

where γ is the surface tension of the liquid and a is the equilibrium droplet radius. This is known as the Rayleigh condition; when the charge exceeds this value the droplet initially stretches into shape similar to a prolate spheroid, before forming two sharp tips from which jets of liquid are ejected. This condition and breakup mechanism has been experimentally verified [14].

Following [4], the charge Q_d on the dust grain can be found from the potential ϕ_d, given by the OML theory, and the vacuum capacitance as

$$Q_d = 4\pi\epsilon_0\phi_d \tag{2.2}$$

and by combining equations 2.1 and 2.2 the Rayleigh condition for the minimum stable droplet size in a plasma can be expressed as

$$a_{min} = \frac{\epsilon_0\phi_d^2}{4\gamma}. \tag{2.3}$$

This result is valid for liquid droplets in typical low temperature industrial plasmas; for a typical plasma-enhanced chemical vapour deposition (PECVD) process $a_{min} = 14.8$ nm, indicating that the final stages of the dispersal process (for $a < 15$ nm) are electrostatically driven. In high temperature plasmas, such as magnetic confinement fusion plasmas where the droplets are molten tungsten or beryllium at over a few 1000 K, thermionic emission of electrons will decrease ϕ_d. This improves the stability of the droplets, but they are still susceptible to electrostatic breakup, as shown in figure 1.

The results from [4] demonstrate that electrostatic disruption is a significant mechanism for destroying liquid droplets in plasma. However, these results are valid only for a stationary droplet in a plasma with no electric or magnetic fields. The first extension to this theory is the inclusion of electric field.

2.2 Inclusion of electric field

In order to understand the behaviour of rain drops in thunderstorms, the breakup of an uncharged droplet in an external electric field was first studied experimentally at least 100 years ago [15] but a valid theory of the disruption was not developed until 1964 by Taylor [16]. The difficulty in describing the stability limit is, in part, due to the fact that a droplet elongates when placed in an electric field, and this deformation makes Rayleigh's linear perturbation theory invalid [17]. Taylor approximated this deformation as a prolate spheroid, and by balancing the electric and surface tension forces at the equator and poles of the

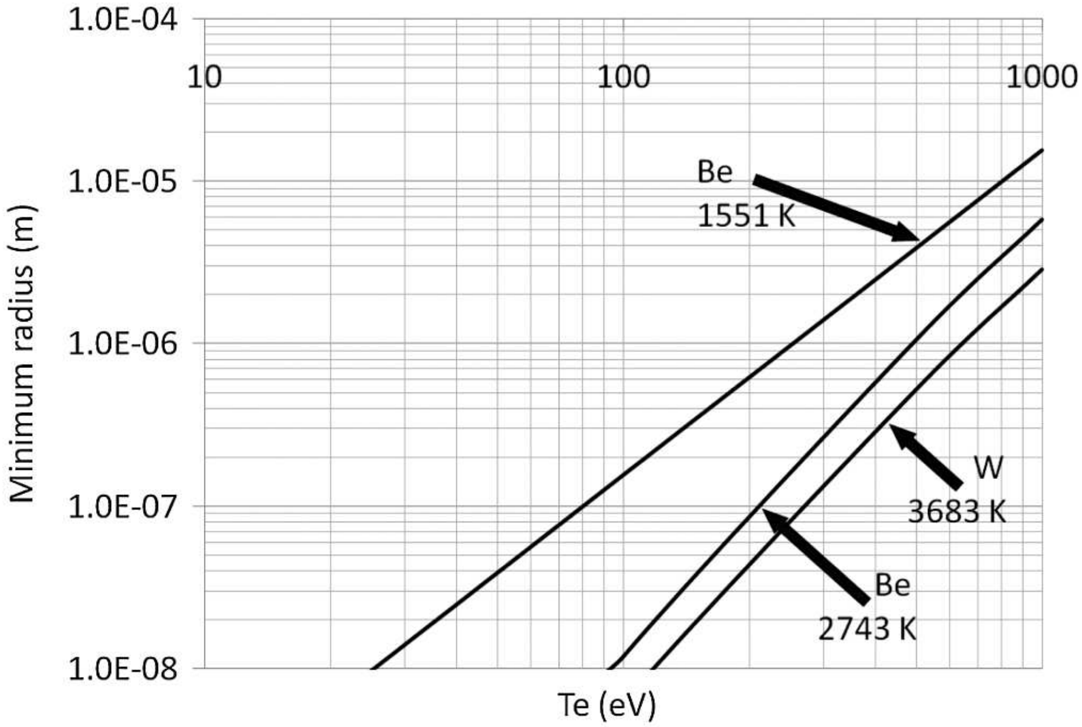

Figure 1: The minimum stable radius of beryllium and tungsten droplets in a fusion plasma as a function of electron temperature in energy units; for conversion to Kelvin 1 eV = 11600 K. Reproduced from [4].

spheroid he found that the droplet becomes unstable for electric fields above the critical value

$$E_c = \frac{c\gamma^{1/2}}{(4\pi\epsilon_0 a)^{1/2}} \tag{2.4}$$

where the fitting constant c was determined by Taylor as 1.625, in agreement with other theoretical [18] and experimental [19] studies. This is known as the Taylor limit.

Taylor's theory is valid for an uncharged droplet, whereas a droplet in a plasma will be charged. Currently the best available study of the stability limit of charged droplets in external fields is a numerical computation by Basaran and Scriven, which balances the electric, surface tension and gravity forces over the free surface of a droplet [20]. Although their results reproduce the Rayleigh and Taylor limits, and agree with experimental studies with charge up to 13% of the Rayleigh limit [21], the intermediate values of critical charge and electric field are evaluated at only three points, requiring some considerable interpolation between values. Despite this, Basaran and Scriven's results can be used to find approximate values for plasma stability.

Electric fields in plasma are particularly strong when the plasma comes into contact with a solid wall, in a boundary layer known as the plasma sheath. The wall becomes negatively charged by the plasma due to the high mobility of electrons compared to ions, in a similar way to the negative charge acquired by dust grains. Attraction of ions to the wall gives the plasma within the sheath a net positive charge, which partially shields the charge on the wall from the bulk plasma over a distance of the order of a Debye length, which is around 1 mm in fusion plasmas. This means there is a large drop in electric potential across the narrow sheath layer, generating large electric fields. Additionally, in many industrial cases the surface in contact with the plasma is a charged electrode, further increasing the electric field strength. It is worth noting that the vessel wall is a key source of dust impurities in a plasma [2, 3], so the sheath region will contain a significant amount of dust and droplets.

In order to investigate droplet stability in the plasma sheath, I have written a code to implement the cold ion sheath model developed by Nitter [22]. This model assumes that the electrons in the sheath have random thermal motion, and are described by a Boltzmann distribution, while the ions flow directly to the wall with no thermal component of motion. Physically this means the ions have a much lower temperature than the electrons; although this is not always true in industrial and fusion plasmas it is good approximation which yields only a slight modification to the observed phenomena. By numerically solving Poisson's equation coupled to the ion equation of motion, the electron and ion number densities, ion velocity and electric potential ϕ were calculated as a function of distance from the wall. The gradient of ϕ was taken to give the electric field strength E. The OML theory for dust grain charging was then applied to find the potential of the dust, ϕ_d, which gives the grain charge Q via equation 2.2. The values of (Q, E) were compared with Basaran and Scriven's results to find the minimum stable droplet radius as a function of distance from the wall, as shown in figure 2.

Figure 2 shows a few interesting features; in particular it can be seen that the electric field decreases the minimum stable droplet radius, meaning that the droplets are more stable when electric field effects are considered. This can be explained by comparing the Rayleigh and Taylor limits. From equation 2.1 it can be seen that $Q_c \propto a^{3/2}$, meaning that large values of droplet charge are stable only above large values of a_{min}. Conversely, equation 2.4 shows that $E_c \propto a^{-1/2}$ and so droplets in large electric fields remain stable above lower values of a_{min}. Hence, the effect of electric fields is to reduce a_{min}. Figure 2 also shows that droplets are more stable close to the wall; this is because electrons in the plasma are repelled from wall, reducing droplet charge and making them more stable.

The theoretical model developed so far describes how electric fields change the stability of a droplet in plasma; this model must be improved to account for magnetic, spin and flow effects and this will be the subject of future work.

2.3 Future work

The easiest extension to the current model is to include a magnetic field, which will change the plasma sheath structure and the OML charging theory. The structure of, and dust grain charging within, magnetised sheaths has been well studied [23, 24] and the charge acquired by the dust was found to be fairly insensitive to the magnetic field. Hence the droplet stability conditions should be similar to before.

Magnetic fields also affect the charging mechanism due to the gyromotion of electrons

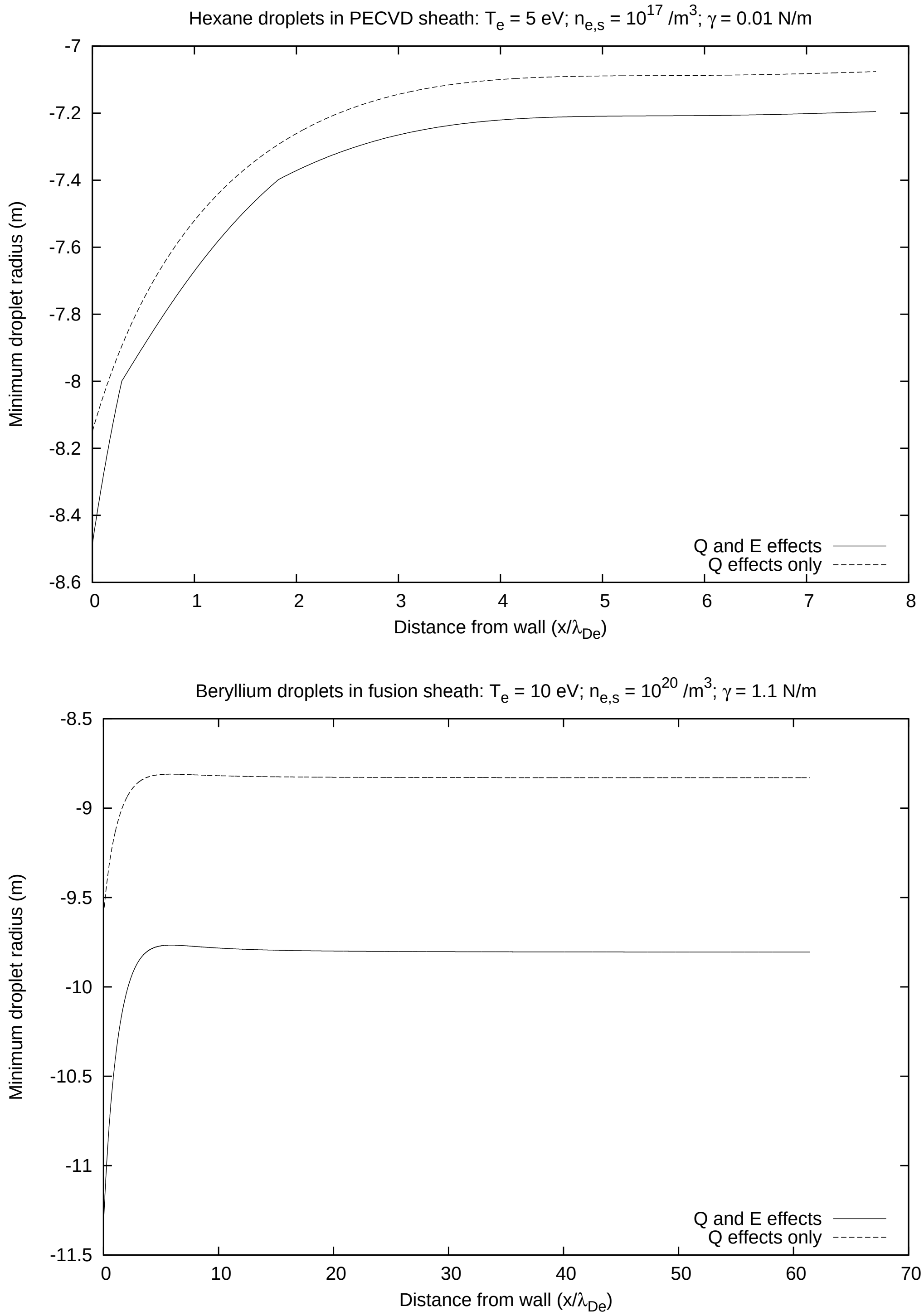

Figure 2: The minimum stable droplet radius for droplets in typical PECVD and fusion plasma sheaths. Droplet stability improves when electric field effects are considered, and droplets close to the wall are shown to be more stable.

and ions in the plasma; this is the circular motion of particles around magnetic field lines at the cyclotron frequency. When ions are collected by the dust grain they transfer angular momentum to the dust, causing it to spin up [25]. The centrifugal force caused by this spinning will change the droplet stability.

The final extension to the stability theory will be for a droplet in a flowing plasma, for example a droplet in a plasma sheath which experiences a strong flow of ions towards the wall. Such flows will impose additional strain on the droplet surface, making the droplet less stable. The problem of droplet stability in a straining liquid flow has already been solved [26] and this theory will be extended to a plasma flow, which requires the force between the droplet surface charge and electrons and ions in the plasma to be considered.

The primary aim of my PhD project is to model these effects, in order to provide a complete theory of droplet stability in a plasma. Subsequent work will consider the translational motion, or dynamics, of a droplet around a plasma.

3 Droplet dynamics

The dynamics of a liquid droplet will be different from a solid dust grain because it can be deformed and its internal motion is damped by viscosity. Usually the largest force on a dust grain is the ion drag force [3], caused by ions in a flowing plasma being collected on the dust grain or interacting with the grain through the Coulomb force [22]. The ion drag force is proportional to the cross-sectional area orthogonal to the plasma flow. For a stable droplet, the factors discussed in section 2 will cause the droplet surface to be deformed, which will change the cross-sectional area of the droplet, and hence change the magnitude of the ion drag force.

Another dynamical effect, as mentioned in section 2.3, is that the gyromotion of ions in magnetic fields causes dust grains to spin up. This spin can be at up to 10^7 s^{-1} for solid dust grains in fusion plasmas, at which point nutation of the rotation axis can change how the dust moves through the plasma [3]. When liquid droplets are spun the viscous shear forces inside the droplet will slow the rotation, reducing the effects of droplet spin compared to a solid grain.

As part of my PhD project it would be interesting to include these dynamical effects, together with the reduced lifetime of droplets due to instabilities, in existing codes for dust transport such as DTOKS [27].

4 Conclusions

This essay has considered the behaviour of micrometre-scale liquid droplets in plasmas, identifying areas to be worked on during my PhD project. In conclusion:

1. Liquid droplets appear in plasmas in a number of important technological uses; they may be injected deliberately into a plasma or be the result of impurities.

2. Electrostatic breakup is a key mechanism for liquid droplet removal.
3. Applying an electric field decreases the minimum droplet radius required for electrostatic breakup, hence fewer droplets are disrupted.
4. Future work will consider the effects of magnetic fields, plasma flow and droplet spin on the conditions for electrostatic breakup.
5. Liquid droplets will have different dynamics to solid dust grains; this should also be investigated.

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