DIRECT ELECTRON-LIQUID ENERGY LOSS SPECTRA MEASUREMENTS USING A LIQUID MICRO-JET

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Abstract. Electron transport through liquids plays a critical role in a number of emerging applications in medicine, particle detectors and nanomaterials. Currently, conducting direct electron-liquid scattering experiments in vacuum conditions poses significant challenges due to the high vapour pressure of liquids and multiple scattering effects. In this work, we present a new electron-liquid scattering experiment that utilises a liquid micro-jet to provide a stable scattering surface for an electron beam under vacuum conditions. Initial measurements are made and compared to existing theory using a Monte Carlo simulation and their limitations are discussed.

1. INTRODUCTION

The application of Low Temperature Plasmas (LTPs) to liquid surfaces forms the basis for a variety of important applications in environmental remediation, medicine and in the synthesis of nanomaterials [Adamovich et al. 2017]. LTPs exhibit typical electron temperatures that are much higher than its equilibrium ion and background neutral species. The resultant heat flux is then much lower when compared to conventional plasmas while maintaining electron energies sufficient to drive reactions within the plasma and into the applied surface [Lieberman and Lichtenberg 2005]. Crucially, this property then allows their application to heat sensitive biological matter under atmospheric conditions. Understanding electron transport across the gas-liquid interface and into the target liquid is thus critical to the application of LTPs to liquid targets and biological matter. In particular, low energy $(< 10 \text{ eV})$ electrons play a key role in driving reactions at the plasma-liquid interface and modelling their transport is critical for the high-level optimisation of efficacy and selectivity of current and future generation plasma-liquid applications.

Theoretical approaches to describe electron scattering through non-polar liquids has recently reached some maturity [Boyle *et al.* 2015], but the same can not be said for polar liquids. Polar liquids exhibit trapped electron states and free electron scattering from spatially, temporally and orientationally correlated dipoles. Given suitable theoretical descriptions of an electron's motion through polar liquids, experimental measurements of electron scattering in liquid environments are also required as a validation step. However, the high vapour pressure and the volatile nature of liquids makes their study under experimental conditions, particularly in vacuums, difficult to achieve [Faubel et al. 1988]. Experiments generally measure either macroscopic properties of electron swarms in liquids [Kubota et al. 1982] or indirectly through photoelectron scattering from liquid beams [Thürmer $et \ al.$ 2013].

Therefore, we have developed a new experiment to measure direct electron-liquid scattering in a vacuum environment through the use of a liquid micro-jet (LμJ). Recently, we presented a feasibility study of such an experiment using a Monte Carlo simulation of model experimental conditions [Muccignat 2022]. A similar proof-ofprinciple experiment was conducted by Nag et al. using an aqueous solution of TRIS (2-Amino-2-(hydroxymethyl)propane-1,3-diol) [Nag et al. 2023]. Here, we present initial results from the experiment and compare them to simulated data and discuss future work.

2. EXPERIMENTAL SET-UP

The scattering chamber utilised here was built originally by Cavanagh and Lohmann [Cavanagh and Lohmann 1999] for coincidence detection of electron ionisation events from a gas beam. In this section, in the interest of brevity, we only detail the modifications made to suit the new LμJ apparatus.

In this work, a LμJ developed by Advanced Microfluidic Systems GmbH¹ replaces the original molecular beam source. An inlet capillary is passed through a Conflat flange into the chamber where it is attached to a crystal nozzle that produces the LμJ. The inner diameter of the nozzle is tapered to produce a liquid beam of a desired size. Liquid samples are prepared in a glass bottle and degassed along with a few grains of salt that are added to reduce electrokinetic charging of the jet during operation [Preissler 2013]. A high pressure dosing pump is then used to draw liquid from the bottle and through the inlet capillary at $0.5 - 1.5$ ml/min.

Figure 1 shows the LμJ with an electron gun and two hemispherical detectors positioned radially around the scattering region. In this experiment, nozzles with diameters of 15, 25 and 40 μm were available and 40 μm was used in this work for its stability. From the nozzle, liquid flows directly downward through the vacuum and into a 1 mm orifice in the centre of the catcher. From there, the liquid drains with gravity and the help of a diaphragm pump where it pools in a vacuum safe bottle submerged in an ice bath awaiting disposal. Both the nozzle and catcher are heated to reduce the risk of the sample freezing. The position of the jet can be externally controlled through two stepping piezo motors along the horizontal axis of the scattering plane. Additionally, the height of the jet can be adjusted manually.

From the catcher, a waste tube removes the expended sample from the chamber through a Conflat flange and into a vacuum safe glass catcher bottle. An additional tube extends from the bottle lid and is attached to a backing pump to prevent backflow into the chamber. Both of these tubes contain valves to allow emptying of the bottle during operation.

¹www.admisys-gmbh.com

Figure 1: Photograph of the present liquid micro-jet apparatus along with labels for each device.

2. 1. RESULTS

During the initial set-up phase of the experiment, a Monte Carlo simulation was used to compare with measured scattering data to align the LμJ within the chamber. Here, we present preliminary measurements of electron-scattering from both the LμJ and from the background vapour.

Using an initial scattering beam of $\approx 250 \text{ eV}$, an energy loss spectra was measured from 251 eV to 220 eV. A strong elastic peak was found followed by a broad and tapering signal that results from inelastic and ionisation scattering processes. In Figure 2 we present the measured energy loss spectra for electrons scattered from the LμJ and compare that to spectra measured when the LμJ was moved outside of the interaction region. For comparison, each spectra is scaled by the maximum value of the liquid spectrum.

In addition, we present simulated spectra using the Monte Carlo simulation for both the liquid and gas scenarios. The simulation utilised the cross-section set proposed by Ness et. al., along with approximate densities and a forward peaked analytic scattering angle function (differential cross-section). Broadly, for the gas density and scattering function selected, the simulation is able to reproduce the elastic peak observed. The most notable difference in each spectra can be found between 230 and 247 eV. In the simulated spectra there exists two small inelastic peaks that are followed by the broad ionisation signal while in the measured data only a single, broad, peak is obvious. Without additional measurements to validate the observed EELS and investigate the dependence upon initial energy and scattering angle, it is difficult to determine the cause for this discrepancy. Rotational processes were not included explicitly in the cross-section set and their inclusion may result in a similar broadening reflected in the simulated spectra. In addition, shifts in the energy loss threshold

Figure 2: Electron energy loss spectra for a 250 eV electron beam incident upon a $40 \mu m$ L_HJ compared to 'gas' phase measurements where the L_HJ was moved to the side. In addition, simulations were conducted using the H_2O cross section set of Ness et al. using an approximate gas phase density and an analytic angular dependence of the differential cross sections. Each pair of liquid and gas spectra were scaled by the maximum value of the liquid spectra for comparison.

for excitation processes, resulting from interaction effects within the liquid, could result in each peak overlapping to more closely represent the measured data. We must note however that these measurements are preliminary by nature and require further investigation and additional measurements to verify.

Overall, we have conducted initial measurements of a new electron-liquid scattering experiment and compared them with existing theory. In future work we aim to leverage the unique nature of this experiment to further our understanding of electron-liquid transport through non-polar liquids.

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