

Toroidal Plasmoid Generation via Extreme Hydrodynamic Shear: Optical and Magnetic Studies

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ABSTRACT

Water is a ubiquitous molecule that mediates our interactions with the world and the dynamics of the universe on scales from astrophysical to biomolecular. Controlling its behavior is an essential step in semiconductor manufacturing and water purification. While commonly viewed as a conductive medium, pure water is in fact quite a good insulator, with a resistivity of $\rho \approx 18 \text{ M}\Omega \text{ cm}$. Such purity is typically short-lived; however, water of sufficient purity allows for unique interactions with solid media as a result of its strong polarization and high dielectric strength ($\epsilon \approx 80$, dipole moment $p \approx 3 \text{ D}$ (Gubskaya and Kusalik, 2002)²). In contact with normal glass, this leads to electric double layer buildup, where the substrate’s superficial atoms—oxygen, typically—preferentially adsorb the polarized hydrogen atoms of the water molecules, resulting in the generation of an electrical potential known as contact electrification. If the fluid is further accelerated against the glass, this effect is amplified via a charge-ripping process whose exact nature is still debated.

In this work, we examine a specific result of this contact electrification and tribo-electrification process via the impingement of a high-speed jet of pure water, normal to dielectric substrates—mainly quartz glass and lithium niobate—with velocities up to 250 m s^{-1} . We observe a velocity-dependent electric field which, at a critical threshold of approximately 120 m s^{-1} , exceeds the breakdown threshold for electrical discharge in air ($\approx 3 \text{ MV m}^{-1}$ (Hong, 2000)), resulting in the formation of a toroidal plasma. We show that the condition of the water, e.g. its resistivity, is essential for this phenomenon, and that the intense shear resulting from this impingement is the primary driver of electrification. By changing the ambient gas to helium or argon, a near-complete shift in the optical spectrum is observed, demonstrating that this is mainly a gas-phase phenomenon.

We further investigate the case of argon, which shows an anomalous pale blue luminescence, and determine that the majority of the visible luminescence in the case of argon is a result of bremsstrahlung radiation, i.e. the deceleration of electrons on impact with argon neutrals, and that a symmetric toroidal core near the jet annulus—functionally a cathode—corresponds to a region of elevated electron temperature within a roughly $10 \text{ }\mu\text{m}$ radial extent of the jet surface. The near-perfect symmetry

²As a frame of reference, a highly polar molecule such as sodium chloride has a dipole moment of approximately 9 D (NIST, n.d.), while a low-polarity molecule such as carbon monoxide has a dipole moment of approximately 0.12 D (Scuseria et al., 1991).

of this interior core, along with the elevated electron energy, suggests that the energy and the symmetry may be related.

Given the non-thermal nature of this plasma, we estimate that a magnetic field of up to approximately 290 mT would be required to balance the nitrogen electron pressure ($n_e k_B T_e \approx 10\text{--}68$ kPa). To determine whether this is phenomenologically feasible, we develop an approach for magnetic field measurement at extreme proximity via remote sensing, in which the sensing elements are directly embedded into the dielectric target. A lab-grown diamond with a thin 5 μm layer of nitrogen-vacancy (NV^-) centers allows magnetometry within approximately 75 μm of the plasma core while requiring no modification to the flow or plasma field. We are able to remotely perform a vector measurement which demonstrates a magnetic field structure consistent with a negatively charged stream of distilled water, and we measure a field strength of up to ± 1 μT at a measurement standoff distance of $z = -75$ μm , which drops rapidly to about 0.25 μT after the onset of plasma. Based on the extensive averaging times of this embodiment, we conjecture that this drop is a consequence of the pulsatile behavior of the discharge, with a signal outside of the dynamic range of our detector.

We then develop a simple electrostatic and magnetostatic picture of the electric field and currents. An electric field model is built from the measurements and the proposed charge distribution. Treating the system as a single closed current circuit, the azimuthal magnetic field follows directly from the enclosed current by Ampère's law, $B_\theta = \mu_0 I / (2\pi r)$, with no field where no net current is enclosed.

We consider two currents within this circuit. A weak fluid charging current, $O(10$ mA) advected with the jet, accounts for the pre-onset field; because our sensor sits *outside* the enclosed current—where an axisymmetric current produces no field—the ± 1 μT we measure reflects the non-axisymmetric part of the distribution. During breakdown a much larger, highly uncertain discharge current (instantaneous peak $O(50$ A), pulsed at a duty cycle $O(10^{-4})$, so its time-average matches the charging current) gives a surface field $O(100$ mT) at the nominal core radius $r \approx 100$ μm , whose magnetic pressure $B_\theta^2 / 2\mu_0$ ($\sim$ a few kPa) acts outward, tending to expand the current loop. This magnetic pressure is of the same order as the computed electron pressures (10–68 kPa)—likely weaker, but not negligible—and both act outward, so a self-consistent model of the observed structure must account for both. While a real azimuthal magnetic field accompanies the flow, the stark symmetry of the plasma core is more plausibly set by the impinging-jet stagnation hydrodynamics

and triboelectric charging than by the magnetic self-field.

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BACKGROUND

Water is perhaps the most well-known, cosmically abundant ³, and vital molecule (Kotwicki, 1991; Westall and Brack, 2018; Van Dishoeck et al., 2014; De Souza Torres and Winter, 2018). Its chemical and mechanical properties result in powerful mediating roles from a range of thermal effects — such as astrophysical cooling or planetary insulation (Yoshida and Gaidos, 2025; Nisini et al., 2010) — to the chemical transport phenomena which enable geophysical and biological activity (Crowley, G  rault, and O’Connell, 2011). Generally, these effects are attributed to its thermal properties, including a high heat capacity and latent heat of phase transitions, or its determination as a “universal” polar chemical solvent. Even if one may be unaware of the technical specificity of these properties, an implicit understanding is almost universal: we use water as a solvent to clean, to brew coffee or tea, and awareness grows of its necessity for industrial manufacturing and data centers on which our reliance is ever increasing.

To whatever degree these processes affect our daily environment, the confluence of these unique properties can ultimately be ascribed to an even more obscure aspect: its electronic properties. A common simplification is to say that water is simply “conductive.” Very pure water, however, (resistivity $\rho \sim 18 \text{ M}\Omega \text{ cm}$) is in fact quite a poor conductor of electricity (Gilson Engineering, n.d.). Only by its polar nature, born of the O-H charge asymmetry illustrated qualitatively in Fig. 0.1(a) and (b), does it readily accept ions into its bulk, resulting in a fluid with meaningful conductivity ($\rho \sim 0.05 \text{ M}\Omega \text{ cm}$ at 10 parts-per-million salt in water). Globally, usage of highly pure water is dominated by semiconductor automotive manufacture, food processing, and processing of pharmaceuticals (*Distilled Water Market Share | industry size & CAGR of 10.57%* n.d.). Despite annual global demand of 6 GL, we still lack a complete understanding of its various interactions with its environment.

In this purified, low conductivity state, the electrostatic field of the water molecule’s electrons is strongly localized. This state is known to exhibit unique interactions with electrons, forming complex variations of the solvated, or hydrated, electron. Fig. 0.1(c) shows an illustration of this type of structure. For additional information on this particular phenomenon, the reader is directed to references Kar and Nair (2025), Katrin R. Siefermann and Abel (2011), Walker (1967). Analogous to exposure to nascent charges, its contact with low-conductivity solid surfaces (such

³In terms of molecules, it is the most abundant *heteronuclear* molecule.

as metal oxides) results in spontaneous polarization due to preferential molecular orientation leading to a bulk potential. This phenomenon, known as contact electrification, is illustrated qualitatively in Fig. 0.1(d) and is most commonly observed at solid-solid interfaces. The resulting polarity depends more exactly on the surface functionalization of the substrate (Custance et al., 2026; Grosjean et al., 2026), but in cases of either $\text{H}_2\text{O} : \text{TiO}_2$ or $\text{H}_2\text{O} : \text{SiO}_2$ contact, the adsorbed molecules induce an effective negative surface charge.

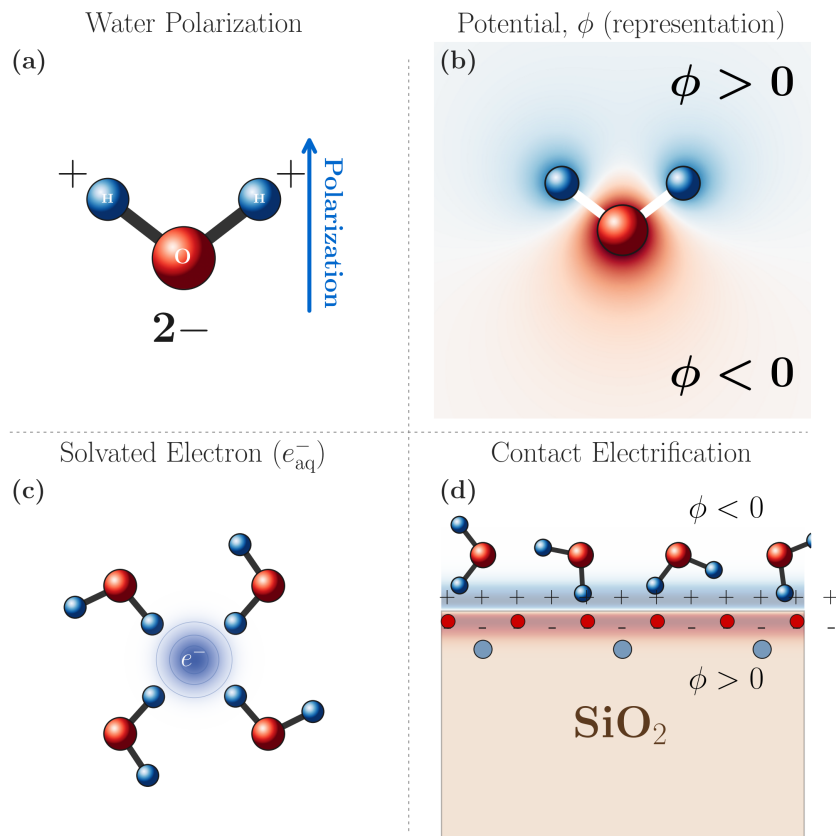


Figure 0.1: Electrostatic charge representation of water molecules (a) in isolation demonstrating the polarization vector, (b) in isolation with a representation of the electrostatic field of equivalent charges, (c) in a circumference around a localized electron, resulting in a solvated electron and (d) generating a contact electrification potential via contact with a silica surface.

In this work, we consider a particular case of contact electrification involving pure water and a dielectric substrate put into relative motion. This amplifies the contact potential and is equivalent to the phenomena known as streaming potential, or tribo-electrification. We look at the case of extreme wall shear generated by an impinging flow of high speed pure water on crystalline oxide substrates, the potential of which

exceeds the breakdown threshold of both the water and surrounding air. Chapter 1 describes the generation of this continuous discharge in air and helium, measuring average plasma parameters from spectroscopic and electrical measurements. Chapter 2 investigates the case of this plasma type in argon, in which its observed color is not apparently congruent with its optical spectra but instead emanates from collisional braking. In Chapter 4, we design a microscope-based magnetometer to remotely measure the near-field structure of this plasma, showing that an axisymmetric azimuthal field exists. In Chapter 5, we develop a simple closed-circuit picture of the electric and magnetic fields: from an electric field model we treat the associated currents through Ampère’s law, with a weak fluid charging current accounting for the field measured before plasma onset and a much larger, highly uncertain (pulsed) discharge current present only during breakdown. The discharge’s self-field exerts an outward magnetic pressure that tends to expand the current loop, of the same order as the intrinsic electron pressure ($n_e k_B T_e$) — likely weaker, but not negligible. Since both act outward, neither confines the plasma alone, and the stark symmetry of the toroidal core is more plausibly set by the impinging-jet hydrodynamics and triboelectric charging than by the magnetic self-field.

*Chapter 1***TOROIDAL PLASMOID GENERATION VIA EXTREME
HYDRODYNAMIC SHEAR**

The following section was adapted with permission for inclusion in this thesis from

M. Gharib^a, S. Mendoza^a, M. Rosenfeld^b, M. Beizai^a, & F.J. Alves Pereira^c, Toroidal plasmoid generation via extreme hydrodynamic shear, Proc. Natl. Acad. Sci. U.S.A. 114 (48) 12657-12662, <https://doi.org/10.1073/pnas.1712717114> (2017).

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1.1 Background

In the epic poem *The Lusiads* (Camões, 1572), the great Portuguese explorer Vasco da Gama narrates how, during his discovery voyage to the Indies, he “saw, and clearly saw, the living light” at the tip of his caravel’s masts. The recount describes from experimental evidence what is known as Saint Elmo’s fire to seafarers facing stormy weather. This luminous phenomenon is, with the more common lightning (Dwyer and Uman, 2014), perhaps the only other atmospheric pressure plasma found in nature. Atmospheric pressure plasmas are a nonthermal nonequilibrium form of plasma that, as a technology, requires no chamber or complex vacuum machinery and has a considerable scientific and industrial potential. Applications in science and engineering span from analytical chemistry, material processing, environmental science, energy, medicine, biology, and low-temperature plasma physics to microelectronics and display technology (Keidar, 2015; Becker, K. H. Schoenbach, and Eden, 2006; Janasek, Franzke, and Manz, 2006; Eden, 2006). Unlike the natural counterparts, these plasmas are obtained by artificially created electromagnetic fields and tend to be nonuniform and unstable (P. Bruggeman and Brandenburg, 2013) without any defining morphology. In this context, miniaturizing and confining plasmas to microscales and in microcavities is considered a viable and promising approach (Becker, K. H. Schoenbach, and Eden, 2006; Janasek, Franzke, and Manz, 2006; P. Bruggeman and Brandenburg, 2013; K. Schoenbach et al., 2008). In air a high potential electric field applied between two electrodes can cause the ionization of the gas elements. The phenomenon manifests as a blue–pink luminescent cloud and is commonly known as an electrical corona. The physics and conditions for onset of this atmospheric pressure plasma are well understood (Loeb, 1965). According to Paschen’s law, $3 \times 10^6 \text{ V m}^{-1}$ are sufficient to trigger a corona in dry air. Here we report on the observation of a naturally formed, stable, unconstrained but topologically confined microscale toroidal plasma in air, at room temperature and atmospheric pressure. We demonstrate the creation of an atmospheric pressure microplasma without external electromagnetic action, through focused hydrodynamic shear. By spectroscopic, electric, physico-chemical, and radio-frequency experimental evidence, we postulate a scenario for its formation. Our study suggests that energetic free electrons are produced by tribocharging from intense hydrodynamic shear in a wall-impinging water jet. Electron collisions with water molecules trigger the formation of charge carriers, allowing the free electrons to reach the water-gas interface and pass into the gas phase, where a plasma forms through the excitation and ionization of the gas elements. When the surrounding gas

is air, the plasma cloud shapes into a topologically coherent structure characterized by a radio-frequency signature compatible with plasma resonance frequencies.

1.2 Results

Discovery

Self-generation of a toroidal microplasma was observed in the flow field of a water microjet in air impinging upon a dielectric surface. The experimental setup consisted of a ruby nozzle with a 100 μm orifice diameter and an air-driven fluid pump, using deionized and bubble-free water. Single crystal quartz (SiO_2) and single crystal lithium niobate (Z-cut LiNbO_3) wafers were used as the target surface, polished or fine ground. A close-up view of the impinging jet is shown in Fig. 1.1. Further details are given in Section B.2.

For jet velocities above 200 m s^{-1} and impingement on a polished quartz wafer, an intriguing bright luminescent blue–pink spot appeared over the core region of the jet. An apparently similar luminous effect has been reported either in passing (Cavaliere et al., 2006) or under different contexts (Miura and Nakayama, 2001; Poulain et al., 2015), yet remained unexplained.

Upon optical magnification, a prominent and rich structure appeared as a well-defined luminescent toroid around the jet core, neighbored by a dark band and followed by a cloud-like glowing region (Fig. 1.1B and Fig. B.4).

The intensity of the luminescent ring was found to increase with jet velocity (Fig. B.1). When the polished surface was replaced by a fine-ground one, a dramatic change occurred in the form of a brighter inner ring and the appearance of radial streamers (Fig. 1.1C and Fig. B.5).

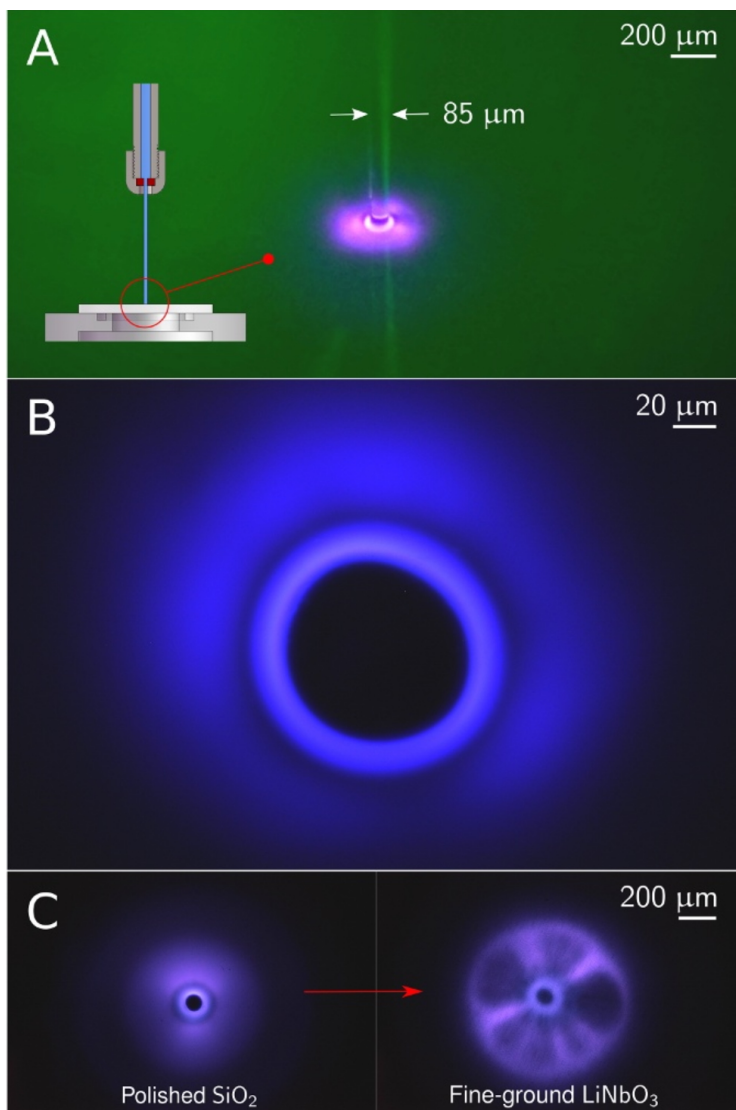


Figure 1.1: Close-up views in the flow field of the impinging jet in air. (A) Photograph of the laminar jet in open air impinging on a polished quartz surface; the jet diameter is $85\ \mu\text{m}$. The transition of the impinging round jet to an expanding radial jet can be seen through the sharp jet annulus. Inset schematic shows the jet in the context of the experimental setup with the nozzle and the wafer support plate. (B) Optical access beneath the experimental setup was used to examine the visual and spectral content of the radiating light through high-resolution microscopy. This photograph, taken with a photographic sensitivity of 640 (ISO standard) and a $1/5\ \text{s}$ exposure time, shows the luminescent structure as observed through the wafer surface with a $50\times$ microscope objective. Water was impinging on a polished quartz surface with a velocity exceeding $200\ \text{m s}^{-1}$. The central dark disk corresponds to the radial cross-section of the jet. (C) Photographs of the luminescent structures taken with a $10\times$ objective, on a polished quartz surface for a jet velocity of $295\ \text{m s}^{-1}$ (Left) and on a fine-ground LiNbO_3 surface for a jet velocity of $212\ \text{m s}^{-1}$ (Right). Streamers, resembling those of common plasma balls, are seen in C, Right as they stretch radially from the impinging site. Note that color levels of images have been enhanced by 33% for better display.

Furthermore, the onset of luminescence was at a significantly lower velocity ($\approx 115 \text{ m s}^{-1}$). Despite the highly dynamic nature of these streamers, giving a visual perception of rotation and resembling the filamentary structures seen in common plasma balls (Cavaliere et al., 2006), the central ring appeared to remain stable (Fig. B.8). Luminescence was not observed when ion-rich water or conducting surfaces were used.

Optical Emission Spectroscopy

To pin down the location of the luminescence, we discharged the jet in an atmosphere of helium on the same polished quartz surface. A change in color confirmed that the light source resided in the gas phase (Fig. 1.2A and Fig. B.6). Changing to a fine-ground LiNbO_3 surface led to similar observations to those with SiO_2 (Fig. 1.2B and Fig. B.7). The confined nature of the toroidal plasma in air also changed to a diffused disk with no clear boundaries or well-defined rings.

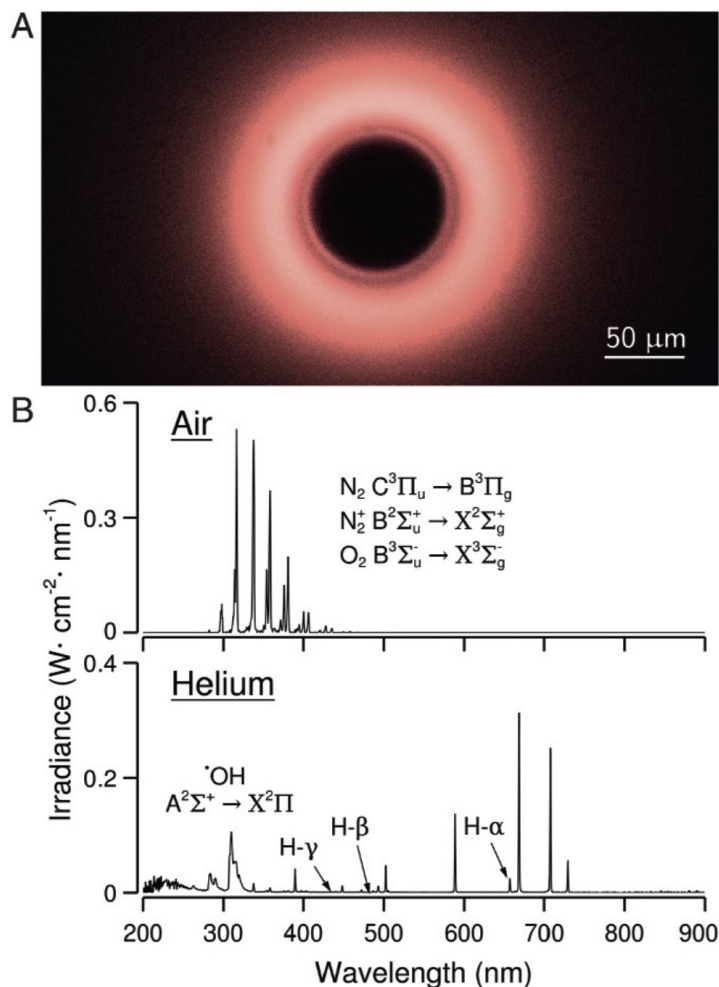


Figure 1.2: Optical emission spectra. (A) The color of the rings changes noticeably when the gas is changed from air to helium. This photograph of the luminescent spot in helium atmosphere was taken through the impinging surface of a polished quartz wafer with a 10 $\times$ microscope objective. Jet velocity was 212 m s^{-1} . (B) Luminescence spectra in air and helium on a polished quartz wafer. In air, we recognize the second positive system of N_2 . In helium, we notice the emission bands of the OH radical in the region 305–310 nm (visible only on SiO_2 , for LiNbO_3 optical transmittance starts at ~ 320 nm) and the first three spectral lines of the Balmer series.

The light spectrum for air (Fig. 1.2B) shows the strong bands of the second positive system of molecular nitrogen (Grum and Costa, 1976) and weaker bands of the first negative system. N_2 is excited and ionized from its ground state ($X^1\Sigma_g^+$) to form N_2^* (state $C^3\Pi_u$) and nitrogen ions (N_2^+)* (state $B^2\Sigma_u^+$).

The spontaneous transition to lower-energy states such as $B^3\Pi_g$ ($N_2\ C \rightarrow B$) and $X^2\Sigma_g^+$ ($N_2^+\ B \rightarrow X$) results in UV emission. These spectral features were also observed with pure nitrogen and are typical of the phenomenon known as electrical corona (Loeb, 1965).

The emission bands of the transition $B^3\Sigma_u^- \rightarrow X^3\Sigma_g^-$ of oxygen are also detected, although strongly shadowed by the N_2 bands. The emission spectrum in helium gas (Fig. 1.2) shows the bands characteristic of the $A^2\Sigma^+ \rightarrow X^2\Pi$ transition of the OH-radical system (OH A–X) in the region 300–310 nm. We also note the presence of the H- α (656.3 nm), H- β (486.1 nm), and H- γ (434.1 nm) lines of the Balmer series of hydrogen.

The presence of $N_2^+\ B \rightarrow X$ transition bands in air and the lack of ionization lines in helium suggest that energies at play can exceed 15.58 eV but are below 24.59 eV, respectively the ionization energies of molecular N_2 and atomic He. Finally, we verified that changing the target surface from SiO_2 to $LiNbO_3$, or from a polished to a fine-ground surface, did not affect the spectral emission pattern both in air and in helium atmospheres.

Plasma Characterization

From the intensity ratio of the OH A $\rightarrow$ X rotational lines (P. J. Bruggeman et al., 2014), we infer a helium gas plasma temperature of $T_g \approx 2100\text{--}2200$ K.

The electron temperature in the helium plasma can be estimated from the peak intensity ratios of the He I lines at 667.8 nm, 706.5 nm, 728.1 nm, and 471.3 nm (Şahin and Tanışlı, 2020; Sasaki, Takamura, and Kato, 1997), yielding $T_e \approx 0.18\text{--}0.22$ eV when helium is the dominant species at atmospheric pressure. This value should be treated as an order-of-magnitude estimate: every published collisional–radiative model for helium targets either fusion conditions ($T_e \approx 10\text{--}100$ eV, $n_e \approx 10^{12}\text{--}10^{14}$ cm $^{-3}$ (Sasaki, Takamura, and Kato, 1997)) or low-pressure regimes that omit radiation trapping, and the only atmospheric-pressure model that treats trapping self-consistently (Chai and Kwon, 2021) still covers $T_e = 2\text{--}5$ eV, $n_e = 10^8\text{--}10^{10}$ cm $^{-3}$ — well outside our regime. The H β Stark-broadening fallback (Gigosos and Cardeñoso, 1996; Surmick and Parigger, 2014) is also unusable: the ~ 1 nm instrument FWHM,

confirmed by the unresolved He I 667.8 and 706.5 nm lines, dominates the ~ 1.1 nm total measured H_β width, leaving only ~ 0.18 nm of Stark broadening buried in the instrument function.

An estimate of the electron density from Stark broadening (Griem and Shen, 1961) of the H- α and H- β lines of hydrogen and from specific lines of atomic He (Griem, Baranger, et al., 1962) (471.3 nm, 501.6 nm, 667.8 nm, and 728.1 nm) is not reliable in this regime: the ~ 1 nm instrument FWHM (see above) dominates the measured line widths, so no quantitative n_e can be extracted for the helium plasma. The large difference between T_e and T_g nevertheless confirms that the helium plasma is nonthermal.

In air, the largely dominant N_2 C $\rightarrow$ B bands of the second positive system are used (Laux et al., 2003) to diagnose a plasma gas temperature in the range of $T_g \approx 1000$ –1500 K. Assuming thermal equilibrium, an electron temperature of $T_e \approx 2.64$ eV is estimated (Britun et al., 2007) from the intensity ratio of the N_2^+ ($B, \nu = 0$) $\rightarrow$ ($X, \nu = 0$) and N_2 ($C, \nu = 0$) $\rightarrow$ ($B, \nu = 5$) transitions, respectively at 391.4 nm and 394.3 nm. Thus, the air plasma is also a low-temperature, nonequilibrium plasma. However, estimation of the electron density from Stark broadening is not possible, for 10.2 eV are needed to put hydrogen into its first excited state.

Finally, we also note that the line ratio between N_2^+ ($B, \nu = 0$) $\rightarrow$ ($X, \nu = 0$) and N_2 ($C, \nu = 0$) $\rightarrow$ ($B, \nu = 0$) or N_2 ($C, \nu = 2$) $\rightarrow$ ($B, \nu = 5$) is strongly related (Paris et al., 2005) to the reduced field strength E/n_n , with E being the electric field strength and n_n the density of neutral particles. We find a mean reduced field strength of $E/n_n \approx 353$ Td, with Td being the Townsend unit.

Electrostatic Field

It is important to appreciate that we observed the toroidal plasma without any externally imposed electric field between well-defined nodes. However, our observation of excitation and ionization spectral lines in air suggests the presence of a strong electrostatic field in the neighborhood of the jet. In fact, a high negative potential with a large dynamic range (-300 to 1000 V) was found between a probe placed inside the plasma and the ground (Section B.2), thus suggesting accrual of negatively charged particles therein. Fig. 1.3 shows the potential field in the close vicinity of the jet. The electric field (arrowed lines) indicates that negative charge carriers should emanate as free electrons from the neck region of the jet, suggesting that this annular area acts as a cathode. Similarly, an anode site is plausibly located in the

radial distance range of 300–500 μm .

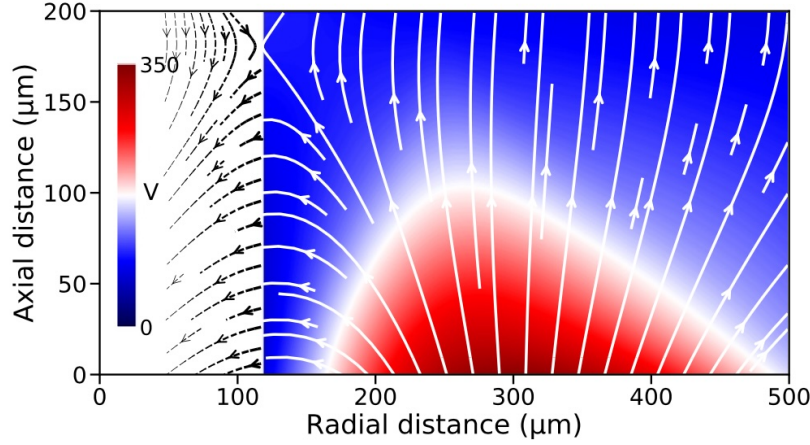


Figure 1.3: Potential and electric fields. We surveyed the region surrounding the toroidal plasma, using a probe at a fixed potential paired with a floating probe placed at various points in the plasma vicinity (Appendix B). The potential V of the interior of the plasma was determined to be consistent enough to act as a reference potential. The color plot shows the isocontour map of the potential field V , after interpolation of the experimental values. The electric field E , represented here as white arrowed lines, was calculated by spatial derivation of the interpolated potential field V . Within 75 μm from the jet boundary where no data are available, the electric field is extrapolated to a radial distance of 50 μm (dashed black arrowed lines).

The air plasma characterization can now be completed (Section B.1) in terms of electron density and plasma resistivity, the latter resulting from electron collisions with ions and neutral particles. Based on the estimated mean reduced field strength $E/n_n \approx 353 \text{ Td}$, and the measured electric field strength $E \approx 10^6 \text{ V m}^{-1}$, we derive a mean neutral gas density $n_n \approx 2.83 \times 10^{18} \text{ cm}^{-3}$.

Based on the electron temperature $T_e \approx 2.64 \text{ eV}$ obtained from the spectroscopic analysis, the electron-ion and electron-neutral collision frequencies are found to be $\nu_{ei} \approx 5.09 \times 10^{11} \text{ s}^{-1}$ and $\nu_{en} \approx 6.56 \times 10^{10} \text{ s}^{-1}$, respectively. Therefore, electron-ion Coulomb collisions dominate over electron-neutral collisions.

The resistivity induced by the electron-ion collisions, or Spitzer resistivity, is $\eta_s = 11.2 \times 10^{-5} \Omega \text{ m}$ and contributes 89% to the total plasma resistivity. The electron density n_e is calculated to be $n_e \approx 1.62 \times 10^{17} \text{ cm}^{-3}$ and the degree of plasma ionization $\alpha = n_i/(n_e + n_i)$ is $\alpha \approx 5.4\%$ under the assumption of quasi-neutrality whereby the ion density is similar to the electron density. These findings are consistent with the observation that Coulomb collisions dominate; however, they

are significantly higher than in typical corona discharges in air and more commensurate with those observed in high-pressure arcs (Huba, 2004) and dielectric barrier discharges (Rajasekaran, Bibinov, and Awakowicz, 2012). The electron density is also substantially higher than that observed in typical corona discharges. Ionization of air constituents other than N_2 , associative and double-ionization reactions, and secondary electron emission mechanisms (Cserfalvi and Mezei, 1996) are suggested causes.

Radio-Frequency Emission

Radio-frequency (RF) measurements were performed using the antenna shown in Fig. B.2. For the highly piezoelectric $LiNbO_3$, the RF power spectrum (Fig. 1.4) shows a strongly discrete content from 3–40 MHz with peaks at 6.8 MHz intervals. This was observed only in the presence of ionized air gas. The peaks are absent with the lesser piezoelectric SiO_2 or when the gas is changed to helium. We also note a broader peak up to about 100 MHz, present for both substrates and dominant in air (e.g. Fig. F.1).

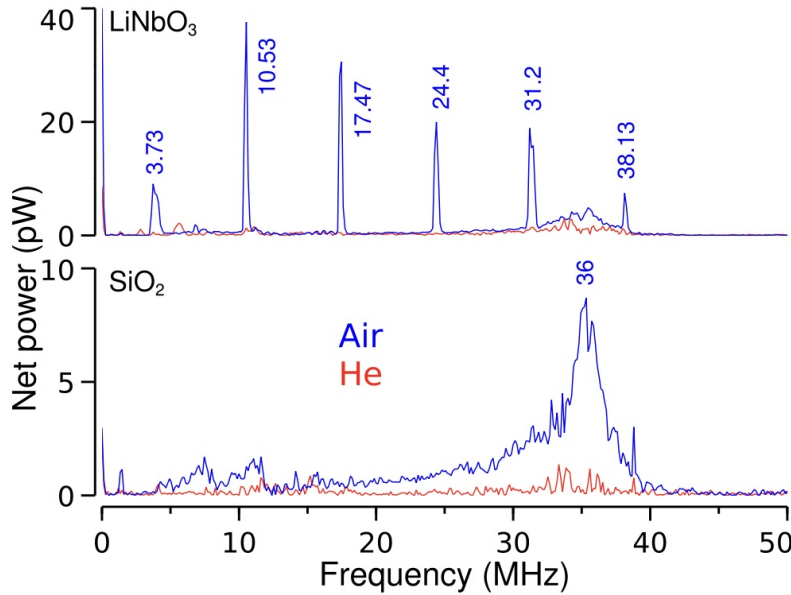


Figure 1.4: RF spectra. Shown are power spectra of the RF signals, on SiO_2 and on $LiNbO_3$, concurrent with luminescence in air (blue line) and helium (red line). Jet velocity was 255 m s^{-1} .

The interaction of electrons with the heavy and positively charged ions is known to induce plasmas to oscillate in response to charge fluctuations (Tonks and Langmuir,

1929). These oscillations, or Langmuir waves, are characterized by a plasma-electron frequency and by a plasma-ion frequency (Section B.1). The latter is also the upper limit of longer-wavelength oscillations known as ion acoustic waves (Fenneman, Raether, and Yamada, 1973), the result of ions and electrons oscillating in phase and producing longitudinal density perturbations similar to sound waves in a gas. We conjecture (Section B.1) that the ≈ 36 MHz peak is likely the signature of an ion acoustic wave instability with wavelength $\lambda \approx 118 \mu\text{m}$, a value in the scale range of the plasmoid documented here. Finally, fluctuations in plasma shape (Tonks, 1931) can induce oscillations distinct from the plasma-electron and plasma-ion oscillations. We suppose that the discrete peaks observed on LiNbO_3 could originate from such plasma shape instabilities, triggered by a possible coupling with the piezoelectric modes of the material or by its pyro- and ferro-electric properties.

Physico-Chemical Characterization of Water

The water was sampled at the edge of the wafer plates. In presence of the air plasma, the pH of water changed from 5.6 to 5.2, respectively before and after corona onset and regardless of the wafer material. This corresponds to a 2.5-fold increase in concentration of cations H^+ from $2.51 \times 10^{-6} \text{ mol L}^{-1}$ to $6.31 \times 10^{-6} \text{ mol L}^{-1}$. At jet speeds below corona onset, the pH remained unaffected with a value close to that of the impinging water.

Air plasmas are known to produce reactive oxygen and nitrogen species (Keidar, 2015; P. Bruggeman and Leys, 2009; Liu et al., 2015), specifically nitrites NO_2^- and nitrates NO_3^- . Assuming the existence of a source of energetic free electrons, several chemical reactions are suggested for our experiment (Table B.1 A–H) that result in the formation of N_2^+ and H_3O^+ ions; the concentration of H_3O^+ defines the pH level. We show (Section B.1) that the pH drops from 5.6 to 5.17 as a consequence of these reactions, a value in close agreement with the 5.2 result from direct pH measurement. Therefore, the air plasma is the cause of water acidification.

At the interface (Table B.1 I–O), bombardment of water molecules by N_2^+ ions accelerated by the electric field induces a secondary mechanism whereby solvated electrons are created via a chemical tunneling reaction (Hart and Anbar, 1970) and released into the gas phase (Section B.1). Generation of near-surface solvated electrons has been reported recently (Rumbach et al., 2015) with an atmospheric pressure air plasma over a water surface. This secondary pathway to electron emission is also a source of $\text{H}^+/\text{H}_3\text{O}^+$ cations, hence affecting further the pH of water,

and a source of $\cdot\text{OH}$ and $\cdot\text{H}$ radicals. No evidence of these latter species was found, suggesting that the free electrons in the gas phase carry less than the ~ 4 eV needed for the first excited state of $\cdot\text{OH}$. This corroborates the 2.64 eV electron energy determined from spectroscopic analysis. Finally, associative ionization of nitrogen atoms (Table B.1B) acts as an important source of N_2^+ ions and free electrons. This reaction is therefore relevant to the interface reactions and can promote both the electron density and the degree of ionization of the plasma.

In the case of the helium plasma, the water pH was found to be unaffected. However, the spectroscopic analysis revealed the presence of the OH radical and several lines of the hydrogen Balmer series (Fig. 1.2B). This suggests that the $\cdot\text{OH}$ and $\cdot\text{H}$ radicals and their ionic counterparts OH^- and H^+ originate from the dissociation of water molecules in the flowing jet or from water vapor near the gas–liquid interface (Table B.1 P–S). For this to occur, a bond-dissociating energy of $493.4 \text{ kJ mol}^{-1}$ is necessary. The water natural autodissociation (Table B.1R) further contributes to the formation of these ionic species. As the cation H^+ bonds with H_2O into H_3O^+ , the latter reverses into H_2O and H^+ and so forth, creating an acidic channel of solvated cations suitable for the transport of electrical current in water. The distinct chemical reactions for the air and for the helium plasmas are energetically congruent (Section B.1), consistent with a common source of energetic free electrons in both cases.

1.3 Discussion and Conclusions

Upon these observations and analyses, we postulate a scenario for the plasma formation in air where the mechanism responsible for the electric field should be closely related to the interaction of the jet with the surface of the dielectric target. One possible candidate is a phenomenon known as streaming potential (Varga and Seymour, 1986), which stems from the triboelectric effect between two nonconducting materials in contact between them and put into relative motion (Ravelo et al., 2011). To verify this hypothesis, we first conducted numerical simulations of the impinging jet flow (Section B.2). The jet is seen to expand abruptly into a radial wall jet (Lienhard, 1995) through a sharp neck (Fig. B.3). The fluid layer then goes through an extreme thinning, reaching a thickness of approximately $3 \mu\text{m}$ at a radius of $617 \mu\text{m}$, followed by a gradual thickening. A highly concentrated region of extreme strain rate ($\approx 5 \times 10^8 \text{ s}^{-1}$) is detected at approximately $66 \mu\text{m}$ from the jet axis and within $1 \mu\text{m}$ from the wall (Fig. 1.5). The wall shear stress at this location reaches about 0.5 MPa. Triboelectric charging is likely to occur in this highly frictional and

concentrated area.

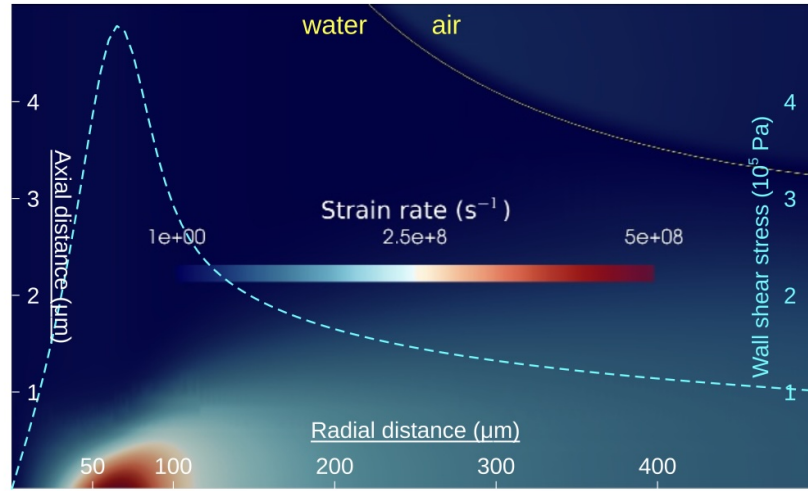


Figure 1.5: Computational fluid dynamics simulation of the impinging jet flow. The color plot represents an area of $500\ \mu\text{m} \times 5\ \mu\text{m}$ and shows the strain rate, with the yellow line outlining the jet interface between liquid water and gas phase. We note the concentrated region of intense strain rate, approximately at $66\ \mu\text{m}$ from the jet axis and within $1\ \mu\text{m}$ from the wall. The cyan curve reports the radial distribution of the wall shear stress (right vertical axis), with a peak of $\sim 0.5\ \text{MPa}$.

Silica-based materials are known to acquire a negative surface charge density when in contact with water (Behrens and Grier, 2001). Therefore, we expect a negatively charged layer on the solid surface side balanced by a positively charged layer on the liquid side (schematic in Fig. 1.6A). As this positive layer is swept by the fast streaming flow, the narrow section of the radially expanding jet populates with positive charges eventually forming an anode site. In the high-shear region, the triboelectric effect between deionized water and the already negatively charged dielectric surface promotes negative charge buildup. Nearest to this high-shear location and with the smallest radius of curvature of the free surface, the annular neck region has a tip-like geometry ideal for charge concentration (Collins et al., 2008), thus forming a cathode site. This anode-cathode configuration is congruent with the electric field topology shown in Fig. 1.3.

We emphasize that this picture does not hinge on the sign of the net charge carried by the *bulk* fluid: whether or not the jet is net-negative, the triboelectric separation of charge at the high-shear interface and the consequent deposition of electrons onto the surface drive the same anode–cathode configuration. The schematic of Fig. 1.6

therefore depicts only the *relative* change in charge — a progressive deposition of electrons onto the surface — and not an absolute charge sign.

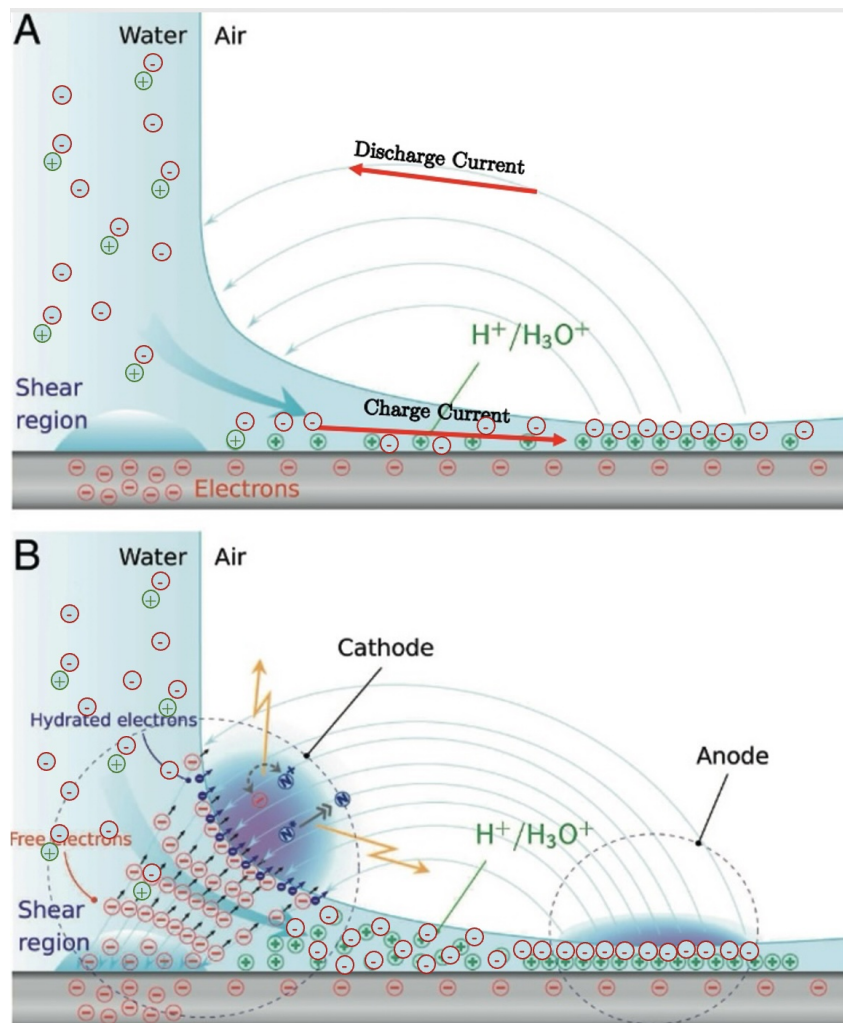


Figure 1.6: Postulated scenario for triboelectric charging and plasma generation in air; the drawing is an illustration only. (A) An early instant of flow over the dielectric wafer: a weak electrostatic field (light blue) forms, with negative charges (red) at the wafer surface and positive H^+ (green) in the water that combine into H_3O^+ and accumulate in the narrow anode region. (B) At higher speed, triboelectric free electrons in the high-shear region dissociate H_2O , raising the water's conductivity so that electrons reach the free surface (the cathode) and pass into the gas, exciting and ionizing it to form the plasma; secondary emission at the air–water interface releases hydrated electrons (blue). The absolute charge of the water is not necessarily known or constant; the schematic depicts only the *relative* change in charge (a progressive deposition of electrons onto the surface), and it is the gradient of the potential—not an absolute charge sign—that is consistent with this charging picture. Straight red arrows indicate the alternation between deposition (charging) and discharge phases; curved blue arrows represent electric field lines.

In this scenario, the free electrons stemming from the high-shear region (Fig. 1.6B) collide and dissociate water molecules in a radiolysis-type process (Schwarz, 1981), producing hydroxyl and hydrogen radicals along with their ionic variants and enabling the transport of current from the high-shear location to the free surface in the annular neck region. The free electrons pass into the air gas phase, ionizing and splitting nitrogen and oxygen molecules, thus triggering the plasma occurrence. Furthermore, ion bombardment of water molecules at the air–water interface creates hydrated electrons, which are then released into the plasma phase. This interface mechanism is intensified by associative ionization reactions introducing additional ions and electrons. As a by-product of these processes, excess cations populate the liquid water phase in the cathode region, causing the acidification of water and eventually driving the transport of current from the substrate to the free surface. We conjecture that the buildup of cations in water inhibits the dissociation of liquid water molecules, halting the formation of hydroxyl and hydrogen radicals. Finally, hydronium cations are also transported toward the anode region where they accumulate to form and sustain the electric potential. To complete the scenario, the free electrons surviving these events travel toward the anode through air along the electric field lines (Fig. 1.6B). Along the path, air molecules are excited and ionized through collisions in a Townsend-type avalanche process forming the air corona. A similar but weaker process occurs at the anode site, denoting the fact that the corona is negative at the cathode and positive at the anode. Evidence of intense spray generation at this location indicates that the positive charge concentration is strong enough to break up the thin liquid sheet into highly charged droplets (Collins et al., 2008; Miljkovic et al., 2013). In the helium plasma case, dissociation of water molecules and excitation of He atoms are likely the only active mechanisms. The absence of He^+ ionization lines indicates that the available electron energies do not reach the 24.59 eV ionization threshold of atomic He, thus preventing the onset of secondary electron emission mechanisms through ion bombardment that would otherwise enhance the electron density of the plasma. The water pH is also not affected as water dissociation is electrically balanced and external sources of cations are absent. Finally, the electrostatic field, plasma, and RF analyses congruently support the idea of a self-induced electromagnetic field as a plausible explanation for the air plasma toroidal topology. Such a condition is normally associated with a self-generated space charge field (Bostick, 1957), which occurs when free electrons and ions permeate the plasma cloud and strongly interact, as evidenced by the electron-ion collisional rate. This situation was verified with ionized nitrogen

only and is supported by an RF signature compatible with plasma oscillations. We suppose that the high electron density and high degree of ionization could also arise from ions being confined by this self-generated electromagnetic field and into a microscale volume.¹ We tested this scenario through electrical grounding near the core region (Section B.2), thus increasing artificially the current density by injecting free electrons into the high-shear region. The test showed an increase in RF intensity and of the electric field strength. We also tested the stability and viability of the toroidal plasma, applying a ± 50 kV potential between the wafer base and the jet nozzle tip. Surprisingly no disruption, damping, or amplification of the plasma structure was observed (Fig. B.9). In summary, we have demonstrated the possibility to create a coherent plasma structure that is both chamberless and unconstrained by external forces, setting a unique paradigm in plasma physics. The study further shows that pure hydrodynamic shear can induce the dissociation of water molecules and the excitation and ionization of gas molecules. Another finding is the possible self-confinement of ions in the form of a toroidal microplasma, without externally applied electromagnetic fields.² This experimental study suggests the discovery of a unique platform to support new experimentation in the broad field of plasma science. The confined and chamberless nature of the plasmoid would allow one to probe the collisional state in greater detail and investigate more effectively the physics of plasma–liquid interactions (P. Bruggeman and Leys, 2009) or of plasma scaling (Janasek, Franzke, and Manz, 2006). The coupling of piezoelectric non-conductive materials with a dielectric liquid through triboelectricity has not been reported to date. In that respect, the degree of accessibility and control offered by our coherent plasma platform can help investigate and probe the triboelectric effect, a still not well-understood phenomenon, or the physics of solvated electrons (Rumbach et al., 2015). Our findings also suggest that exotic properties such as pyro-electricity (Brownridge and Shafroth, 2001) and ferro-electricity (Rosenman et al., 2000) could be explored to enhance the energy range of our plasma process and inspire new concepts of plasma devices for material processing or for the emerging field of plasma medicine (Yan et al., 2015). Our platform could also be of interest within the technological challenge of plasma self-confinement (Binderbauer et al., 2015) or suggest pathways to create energy-storing coherent plasma structures at atmospheric pressure (Smirnov, 1993).

¹The analysis of Chapter 5 shows that the azimuthal self-field exerts an *outward* (loop-expanding) magnetic pressure; it does not provide magnetic confinement. The toroidal symmetry is more plausibly set by impinging-jet stagnation hydrodynamics.

²See the preceding footnote and Chapter 5.

1.4 Summary

Saint Elmo's fire and lightning are two known forms of naturally occurring atmospheric pressure plasmas. As a technology, non-thermal plasmas are induced from artificially created electromagnetic or electrostatic fields. Here we report the observation of arguably a unique case of such a naturally formed plasma, created in air at room temperature without external electromagnetic action, by impinging a high-speed microjet of deionized water on a dielectric solid surface. We demonstrate that triboelectrification from extreme and focused hydrodynamic shear is the driving mechanism for the generation of energetic free electrons. Air ionization results in a plasma that, unlike the general family, is topologically well defined in the form of a coherent toroidal structure. Possibly confined through its self-induced electromagnetic field,³ this plasmoid is shown to emit strong luminescence and discrete-frequency radio waves. Our experimental study suggests the discovery of a unique platform to support experimentation in low-temperature plasma science.

³Revised in Chapter 5: the magnetic self-pressure is directed outward; the toroidal symmetry is attributed to hydrodynamic stagnation flow.

*Chapter 2***BREMSSTRAHLUNG RADIATION FROM TOROIDAL
PLASMAS GENERATED THROUGH HYDRODYNAMIC
SHEAR**

The following section was adapted for inclusion in this thesis with permission from

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2.1 Introduction

Toroidal Plasma in Air and Helium

A detailed study of this plasma in air and helium, and specifics of the hydrodynamic system underlying this class of hydrodynamically generated plasmas can be found in (Gharib et al., 2017). Here, a brief description is provided.

A high-pressure jet of deionized water ($\varnothing = 85 \mu\text{m}$) impinges upon a dielectric substrate. The resulting water film spreads radially, forming an axisymmetric liquid sheet within which the wall shear stress can exceed 1 MPa in a thin toroidal region. Near the impingement point, the triboelectric effect causes electrons to be adsorbed to the substrate's surface, forming a polarized interface (see (Lin et al., 2013; Ravelo et al., 2011), for additional examples and discussion of triboelectric charging of water-solid interfaces). With the film's radial spread, cations are advected downstream, as previously confirmed by electrical and pH measurements, forming an anode as a counter-electrode. When the electric field is sufficiently strong, breakdown of the insulating film and gas layers occurs, leading to the ejection of electrons into the air in a cascade-type discharge, forming the toroidal plasma.

The critical shear required to initiate this phenomenon is highly dependent on the interfacial characteristics, predominantly surface roughness of the target material. On rougher surfaces ($Ra \sim 1 \mu\text{m}$), the plasma initializes at a low jet velocity ($\sim 100\text{--}150 \text{ m s}^{-1}$), while on smooth surfaces ($Ra \sim 1 \text{ nm}$) a higher velocity is required ($> 150 \text{ m s}^{-1}$) (Gharib et al., 2017). Secondarily, impingement on a rough surface typically produces a plasma with fixed radial streamers, dependent on the precise impingement location. On smooth surfaces, the plasma might assume a symmetric configuration as in Fig. 2.2, or one in which streamers appear that rotate about the jet axis, as streamer origins are not locked to features of the surface roughness. See supporting information of Gharib et al. (2017) for videos demonstrating these phenomena.

In both cases, the electron energy is sufficient to ionize nitrogen, as confirmed by the spectral emissions and the violet appearance of the nitrogen plasma, with an ionization fraction estimated by Gharib et al. (2017) of up to 5%. Notably, this is higher than in typical corona discharges, but similar to high pressure arcs and dielectric barrier discharges. Suggested causes are associative and double-ionization processes, and secondary electron emission. In a helium ambient, neutral excited states appear in the optical spectrum, corresponding to an orange appearance of the helium plasma in which the ionization fraction is less than 0.1%. In both cases,

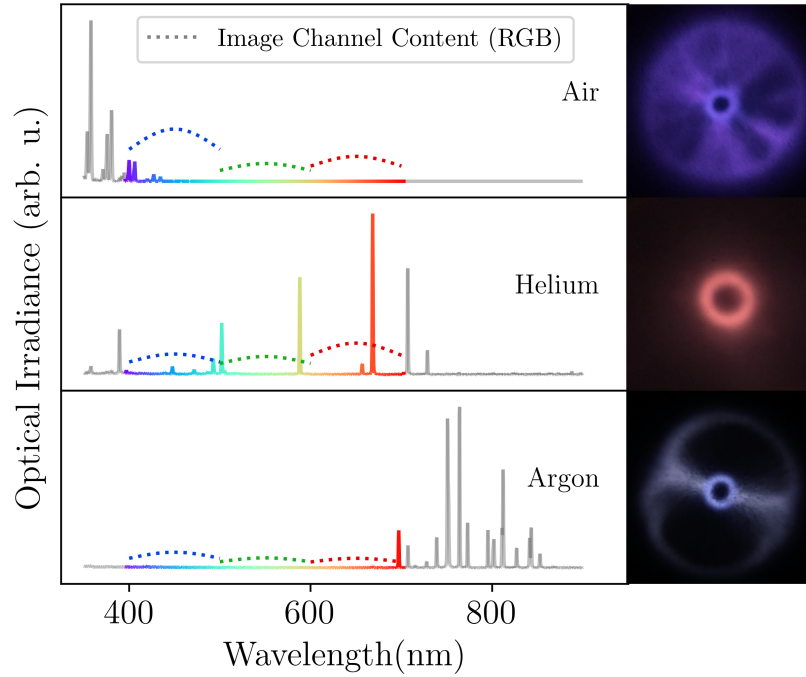


Figure 2.1: Overview of optical spectra, images, and a spectral representation of the RGB content of the images of the toroidal plasma in air (top), helium (middle), and argon (bottom)

the plasma appearance agrees well with the optical spectral content. A projection of the RGB content of their images onto the visible spectrum (shown in Fig. 2.1) indicates good agreement between the light captured by photos and the empirical spectral data.

Toroidal Plasma in Argon

Argon plasmas can exhibit increased conductivity in relation to those in helium, but the electric field distribution in both cases has been shown to be nearly identical, stretched or compressed spatially as a result of differing mass ratios (Mansour and El-Sayed, 2013). Thus, the underlying physics that *generates* these plasmas is comparable, but ultimately the variable plasma behavior is emergent from properties of the gas phase, such as mass, ionization energy, and energetic degrees of freedom. In argon, such as in helium, we expect a low degree of ionization, low electron density, and substantial luminescence from neutral atom excitation (the Ar I system).

This Ar I excitation system comprises mainly infrared optical content (690–950 nm). In contrast, the excitation system of singly ionized argon (the Ar II system) comprises visible optical transitions (400–500 nm). The ionization energy of argon (15.76 eV) is relatively high, but within the energy range available in the air and helium-based systems, and may be made further accessible by argon penning excitation, in which argon metastables act as a gain medium to produce excited states which electrons alone cannot (Sahin et al., 2010). These metastables can lower the atmospheric breakdown voltage and change discharge characteristics (Yoshinaga and Akashi, 2013), leading to an ion density twice that of electron density under atmospheric conditions (He et al., 2013). Ultimately, contributions from the ionized system must be considered not only with respect to energy, but also to both the density and transition probabilities of the species.

Operation of the plasma under argon revealed significant visible light, seen as bluish-white both to the naked eye and in images, as shown in Fig. 2.2. In these images, a bright blue toroidal ring encircles the dark core of the water jet. Outside this ring, the plasma transitions to a diffusive bluish-white, indicative of broad spectral content. Additional examples of this phenomenon are shown in the Gallery (images ArQ1–ArQ4 and ArL1–ArL4). While the blue toroidal ring might indicate Ar II transitions, akin to those in argon ion lasers, the white component in the external regions suggests the involvement of additional processes across the optical spectrum (Kelly et al., 2016). To elucidate the underlying mechanisms in this toroidal argon plasma, we turn to optical emission spectroscopy (OES) as a straightforward, accurate, and effective method for identifying excited states. In this work, we analyze characteristic emissions from the Ar I and Ar II systems, as well as spectral continua, to determine what underlying mechanisms are congruent with the distinct character of the observed argon plasma. Through a multispectral imaging approach, we spatially resolve the T_e and n_e of these excitation systems to distinguish plasma phases between the inner toroidal ring and the outer diffusive region of the plasma.

2.2 Experimental Setup

Plasma Generation

The experimental apparatus used is described in-depth in the previous work. Fig. 2.3 shows a schematic (a) and photo (b) of the setup, and an image of the argon plasma as viewed through the quartz window (c). This image, and all subsequent measurements and images hereafter were performed on a Z-cut lithium niobate substrate from Precision Micro-Optics, Inc., with a surface roughness of $Ra \sim 10 \mu\text{m}$.

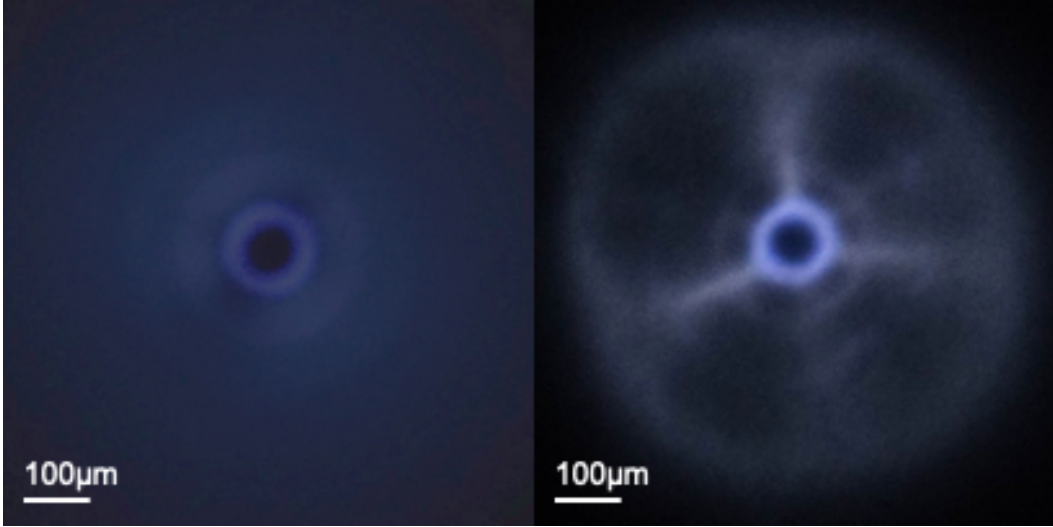


Figure 2.2: Images of toroidal argon plasma on a polished quartz surface (left), and a rough quartz surface (right).

Imaging and Spectroscopy

Optical emission spectroscopy was performed using a B&W Tek spectrometer (BRC112EU) calibrated *in situ* with a NIST-traceable source lamp (Oriel 45W Halogen, 63358).

A Sony A7S camera adapted to an Olympus semi-apochromatic objective (LUC-PLFLN20X) was used for all imaging. Photos were taken as raw images at the camera's native ISO (ISO 100).

For multispectral imaging, green and red optical bands were defined as $B_g = \{\lambda \mid 500 \text{ nm} \leq \lambda \leq 515 \text{ nm}\}$ and $B_r = \{\lambda \mid 618 \text{ nm} \leq \lambda \leq 632 \text{ nm}\}$, matching the set of chosen interference filters (Thorlabs FBH515-10 and Thorlabs FLH633-5). An exposure time of 4 seconds was used for narrow-band imaging.

For processing, the blue and red channels' data were extracted from the raw images. These channels were chosen to represent the green-light and red-light measurements, respectively, as the camera's filter and sensor characteristics are most similar between the blue and red channels at the chosen wavelengths. Prior to analysis, an alignment was performed on the image pairs to adjust for any incidental in-plane translation.

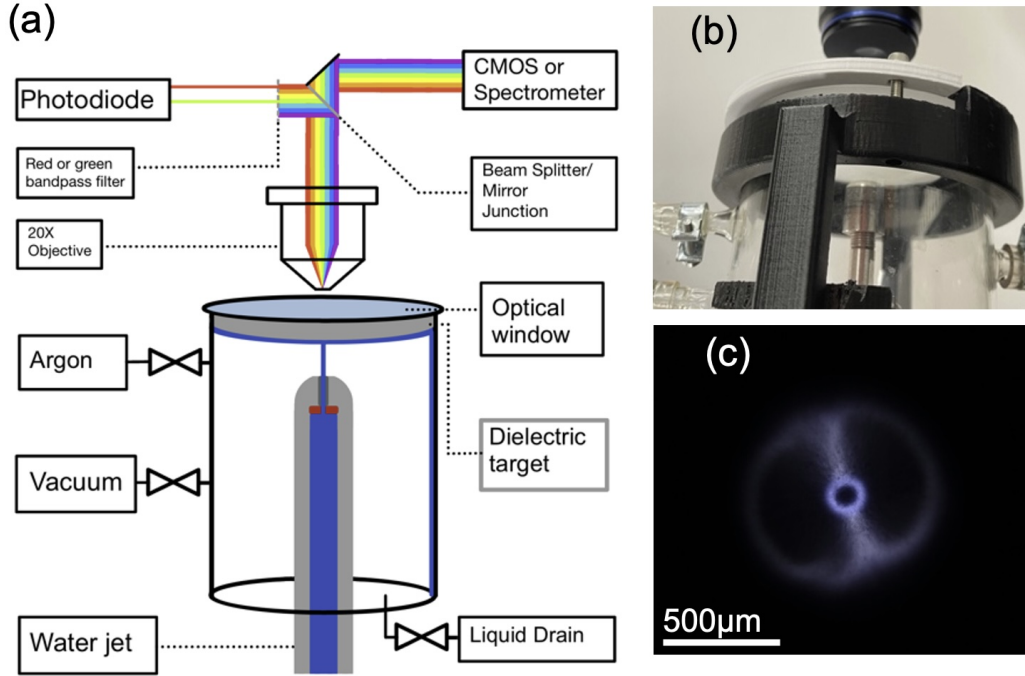


Figure 2.3: (a) Schematic of the experimental setup. (b) Photo of the plasma chamber and Laowa lens. (c) Image of toroidal plasma in an atmospheric pressure argon ambient, showing an inner toroidal ring and diffusive outer region.

2.3 Results and Discussion

Emission Characteristics

Fig. 2.4(top) shows the calibrated emission spectrum with and without magnification. Reference lines of the Ar I and Ar II systems (reported by Schulze et al. (2008) and Bridges (1964)) are shown for comparison. Without magnification, only infrared spectral lines are apparent. In the magnified spectrum Fig. 2.4(bottom) it becomes evident that the visible band ($\sim 400\text{--}700\text{ nm}$) contains broad spectral content, taking the form of a violet-biased continuum with some weak spectral lines. Data are made available by Mendoza (2024).

Radiative Processes

Comparison of the measured spectrum against the Ar I and Ar II signatures demonstrates that both systems are present, but that the Ar I population is substantial in comparison to the Ar II system. In the visible band, an irradiance continuum of similar maximum intensity to the Ar II lines underlies both systems.

In an atmospheric plasma where argon and water are present, continua (ϵ_c) from

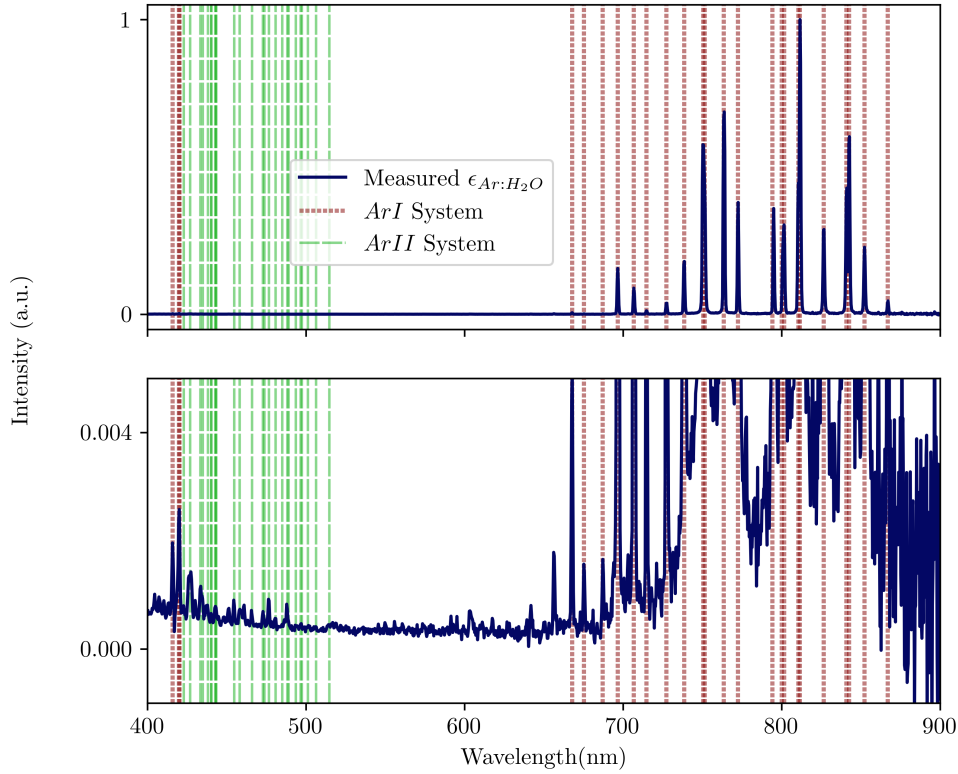


Figure 2.4: Optical spectra of argon plasma from 400–900 nm shown in full scale, and magnified 250x. The strongest spectral lines of the Ar I and Ar II excitation systems are shown for comparison.

both bremsstrahlung radiation and hydrogen dissociation (ϵ_H) are possible. Above 450 nm, ϵ_H is weak (S. Park et al., 2015). To confirm this, the jet was operated with D₂O to instead target the deuterium dissociation continuum (ϵ_D , shown in Fig. 2.5). Any variation between ϵ_D and ϵ_H in this region remained below the noise floor of the detector, confirming that these continua have a negligible influence on our consideration of the visible spectrum. Therefore the primary source of ϵ_c must be a form of bremsstrahlung radiation, which occurs during the deflection of charged particles, such as electrons when impacting heavy ions or neutral atoms.

Bremsstrahlung can comprise electron-neutral (ϵ_{ea}) and electron-ion (recombinant ϵ_{ei}^{fb} and non-recombinant ϵ_{ei}^{ff}) types. Although evidence of Ar II lines are present, and its species are likely increased due to the gain effect of Penning excitation, the

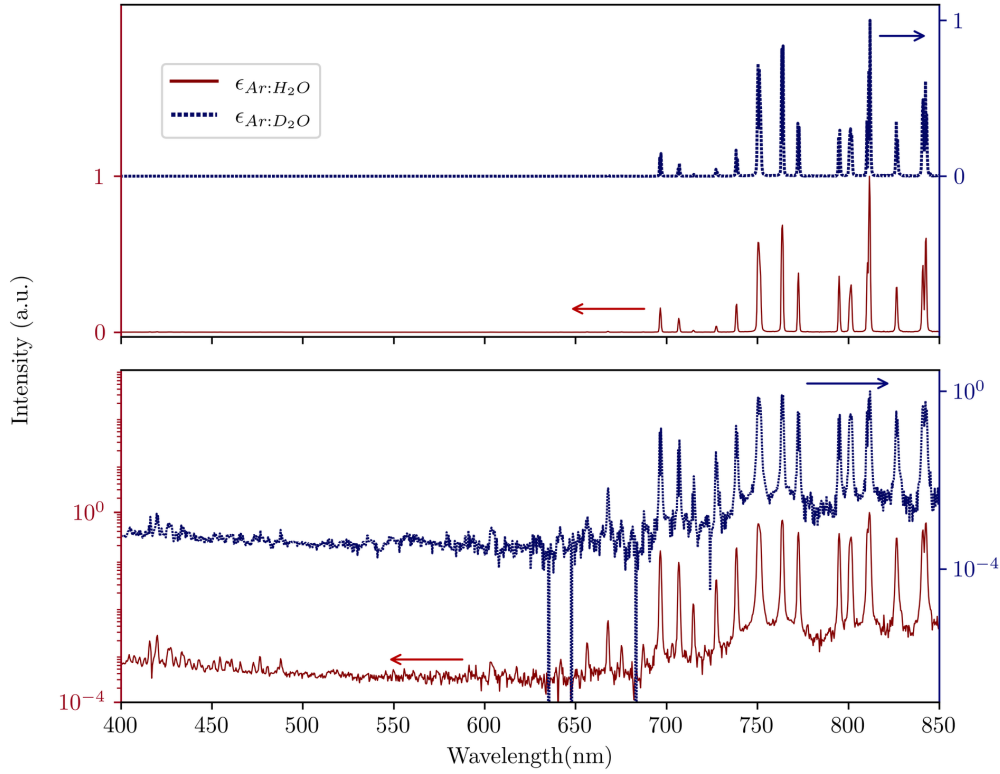


Figure 2.5: Optical spectra of argon plasma originated from H₂O (red) and D₂O (blue) jets, with focus on characteristic peaks (top) and magnified to show the respective continua (bottom).

neutral atom density is several orders greater than the ion density (Y. Fu et al., 2018) at atmospheric pressure. This is also reflected in the relatively low electron density, $n_e \approx 2 \times 10^{15} \text{ cm}^{-3}$, estimated below. Thus the rate of electron-neutral collisions is also much greater than that of electron-ion collisions for the argon plasma. This is consistent with the well-documented ability of argon discharges to sustain stable, diffusive plasmas at lower electron densities and applied voltages than molecular gases such as nitrogen (Staack et al., 2008; Massines et al., 2012; Raizer, 1991); nitrogen plasmas, by contrast, require greater localized energy input to maintain a comparable discharge. Therefore, the primary form of bremsstrahlung is electron-neutral bremsstrahlung radiation ϵ_{ea} (Hundley, 1962). The respective energy content associated with this continuum and the discrete excitation sources ($\epsilon_{\text{Ar I}}$, $\epsilon_{\text{Ar II}}$) was determined by numerical integration of the spectrum.

These energies, calculated as normalized quantities ϵ^n , are tabulated in Table 2.1. From this, we see that in water, the Ar II system contributes $\sim 0.1\%$ to the total irradiance while ϵ_{ea} contributes $\sim 1\%$. The Ar : D₂O case is similar, except that the Ar II content is lessened. The similarity between ϵ_{ea} in both cases further supports that hydrogen and deuterium dissociation continua contribute negligibly to visible radiation. As a result, we confirm that the primary source of ϵ_c in this system, and therefore of visible light, is electron-neutral bremsstrahlung (ϵ_{ea}).

Bremsstrahlung Simulation

Bremsstrahlung, (“braking”) radiation, is a specific form of Compton scattering (Bochek, Deiev, and Maslov, 2012) which occurs to a small degree in nearly any process with electron scattering, and has been considered as a mechanism of sonoluminescence (Frommhold, 1998). It can be used to deduce temperature distributions for Maxwellian systems (Lamoureux et al., 2000) and is applied to systems of hot electrons, where its contribution is common.

To determine the electron temperature characteristic of ϵ_{ea} in this plasma, we prepared a synthetic bremsstrahlung spectrum to fit the OES data. In the regime where ϵ_{ea} is the dominant continuum, the emission profile is given by S. Park et al. (2015):

$$\epsilon_{\text{ea}}(\lambda, T_e) = \frac{(n_a n_e)}{(\kappa T_e)^{3/2}} \sqrt{\frac{2}{\lambda m_e}} I(T_e, \lambda), \quad (2.1)$$

where $I(T_e, \lambda)$ is the integral:

$$I(T_e, \lambda) \equiv \int_{\kappa T_e}^{\infty} (\kappa T_e)^2 Q_{\text{ea}}^{\text{mom}} \left(1 - \frac{ch}{2\lambda \kappa T_e} \right) \sqrt{1 - \frac{ch}{\lambda \kappa T_e}} d(\kappa T_e), \quad (2.2)$$

Table 2.1: Irradiance from the primary excitation systems of the plasma normalized to irradiance from the Ar I : H₂O system, presented for both the Ar : H₂O and Ar : D₂O cases.

Emission Source ¹	Ar : H ₂ O	Ar : D ₂ O
$\epsilon_{\text{Ar I}}^n$	~ 1	~ 0.9
$\epsilon_{\text{Ar II}}^n$	$\sim 10^{-3}$	$\sim 5 \times 10^{-4}$
ϵ_{ea}^n	$\sim 10^{-2}$	$\sim 10^{-2}$

and $Q_{\text{ea}}^{\text{mom}}$ is the argon momentum transfer cross-section, experimentally determined by M. M. Popović, S. S. Popović, and Vuković (1974) to be approximated well by:

$$Q_{\text{ea}}^{\text{mom}} = 8.05e^{-28.024\kappa T_e} + 0.99 (\kappa T_e)^{1.743} e^{-0.136\kappa T_e}, \quad (2.3)$$

Here, ϵ , T_e , λ , h , c , κ , n_a , n_e , and m_e are the emissivity, electron temperature, emission wavelength, Planck's constant, speed of light, Boltzmann's constant, neutral density, electron density, and electron mass, respectively.

Park, et al. demonstrated that the function $\epsilon_{\text{ea}}(\lambda, T_e)$ could be fully determined by measuring ϵ_{ea} at only two wavelengths. Thus, we define the emissivity ratio as that from variable optical bands:

$$\mathcal{R}(T_e) = \frac{\int_{B_g} \epsilon(\lambda, T_e) d\lambda}{\int_{B_r} \epsilon(\lambda, T_e) d\lambda}. \quad (2.4)$$

Synthetic bremsstrahlung continua were generated in the T_e range of $0.5 \text{ eV} < T_e < 15 \text{ eV}$. Their associated $\mathcal{R}$ was then calculated using the bands B_g and B_r , matching our optical filters. Note that the choice of optical bands eliminates discrete spectral content from hydrous species, Ar I transitions, and Ar II transitions, as demonstrated by S. H. Park et al. (2014). With this synthetic data, a monotonic interpolation function representing Eq. 2.4 was generated to calculate T_e from such optical band measurements.

Electron Temperature

The mean bremsstrahlung electron temperature based on OES data was calculated to be $T_e \sim 1.1 \text{ eV}$. The emission spectrum, data from $B_g \cup B_r$ used for modeling, and interpolated and extrapolated bremsstrahlung profiles are shown in Fig. 2.6. Note that the choice of B_g and B_r removes the majority of discrete spectral content from this measurement. Spectral lines which may give rise to contamination in these bands are primarily the Ar II lines at 501 nm, 506 nm, and 514 nm, as noted in Fig. 2.4. However, the population of these species is lower than that of Ar I, and their transition rates are about five orders lower as well (Dunn and Ross, 1976). It should be noted further that highlighted data presented in Fig. 2.6 are barely above the noise floor of the spectrometer, and thus the appearance of small peaks in these bands cannot with certainty be attributed to spectral transitions. Nevertheless, the

discrete contribution to these bands may be up to 5% of the total emission, and thus may contribute to the uncertainty in the T_e calculation.

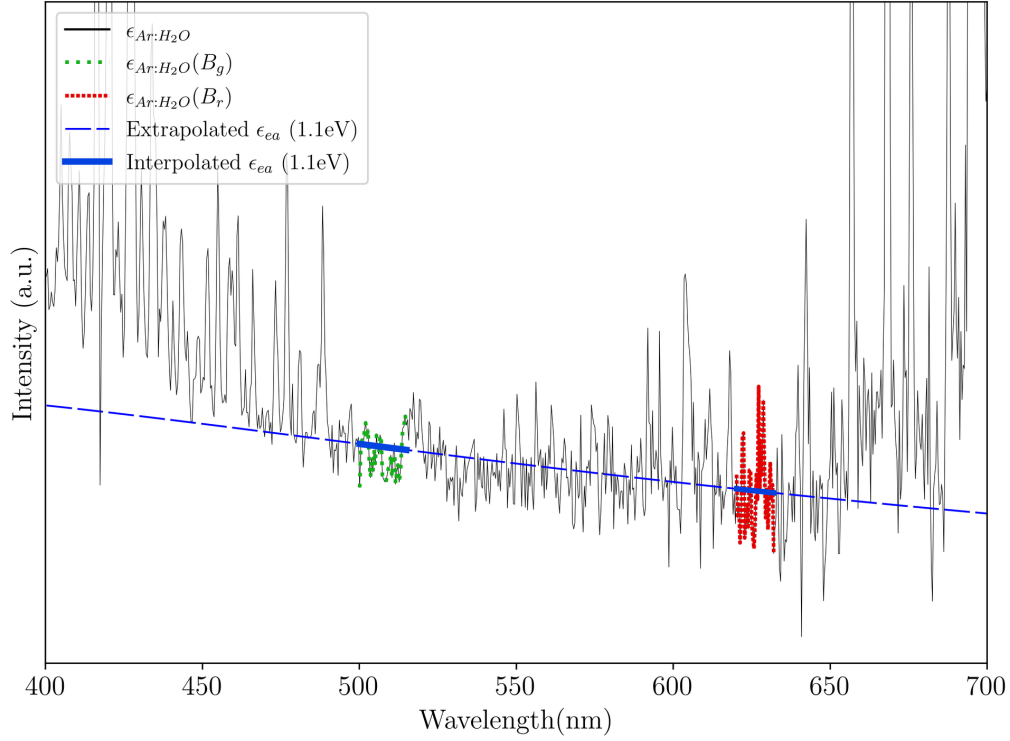


Figure 2.6: Calibrated OES data from the argon plasma (black), data used for modeling (green and red), interpolated ϵ_{ea} (solid blue), and extrapolated ϵ_{ea} (dashed blue) profiles.

To spatially resolve T_e , images were taken within the B_g and B_r bands, each having an uncorrelated error of $\epsilon \sim \pm 5\%$ (Image data is made available at Mendoza (2024)). Their pixel-wise ratio is then the array $\mathcal{R}$, with resulting error $\pm 7\%$.

After processing the images through the mapping $\mathcal{R} \rightarrow T_e$, an array of T_e is produced. Fig. 2.7 shows the visible-spectrum image of the plasma (Fig. 2.7a), B_g image (Fig. 2.7b), B_r image (Fig. 2.7c), and T_e as a heatmap (Fig. 2.7d). Error in T_e shows reasonable sensitivity below ~ 2 eV, but large errors near its end range, due to the non-linear nature of the T_e mapping. Due to the low intensity of red light from B_r , the corresponding image was amplified for clarity.

In the original image, the toroidal plasma appears with an inner blue ring from which two streamers emanate. These streamers expand in breadth, connecting at

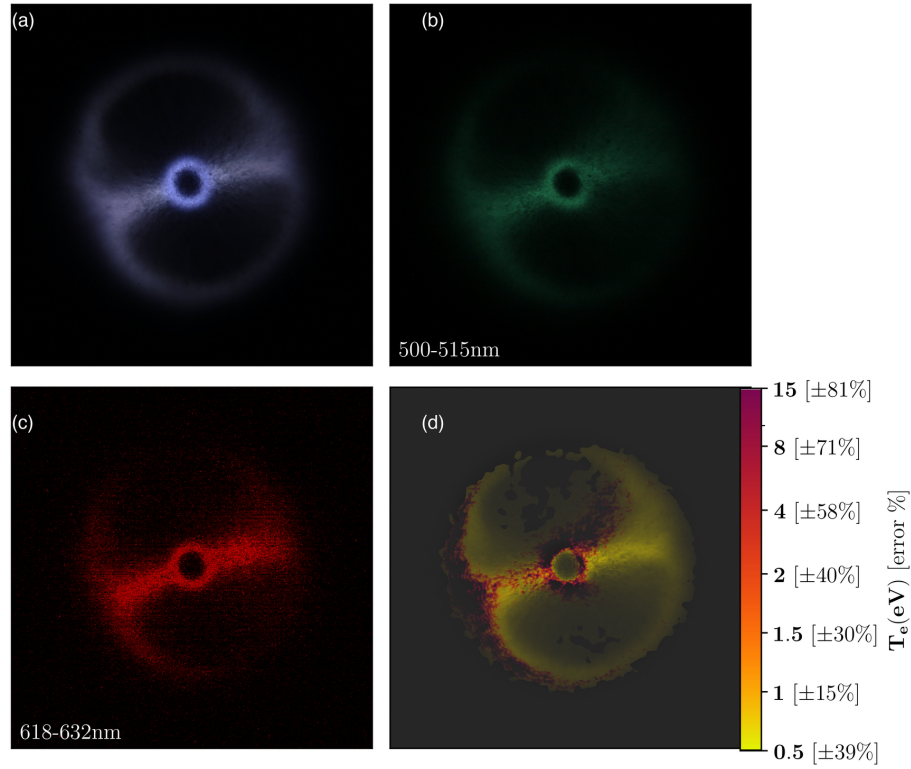


Figure 2.7: Photographs of the toroidal plasma in argon formed on a rough lithium niobate substrate ($Ra \sim 10 \mu\text{m}$), (a) unfiltered, (b) filtered to B_g , and (c) filtered to B_r . Estimated T_e corresponding to the electron-neutral bremsstrahlung process (d).

a second outer radius. Fig. 2.8 displays the relevant regions for discussion: inner ring, outer ring, overestimation regions, and jet core.

The streamer morphology, influenced by intrinsic patterns in the roughness of the lithium niobate substrate and the precise impingement location, is clearly visible in both the green and red channel images, as well as in the T_e map. The T_e map shows that the right streamer has a slightly lower temperature (0.5–1 eV). In contrast, the left streamer displays sporadic spots of higher temperature, with some saturation. Toward the left streamer border, the elevated T_e is likely due to a shift in the impingement site, resulting in a shorter streamer and an overestimated $\mathcal{R}$ near the plasma edges, which in turn inflates T_e . This edge effect is also noticeable at the inner plasma border, where minor asymmetries appear between the right and left interior borders of the inner toroidal ring. Because the plasma geometry (size, shape, and both number and angle of streamers) is closely tied to surface roughness, we conjecture that the asymmetries in the streamers are due to asymmetries in this

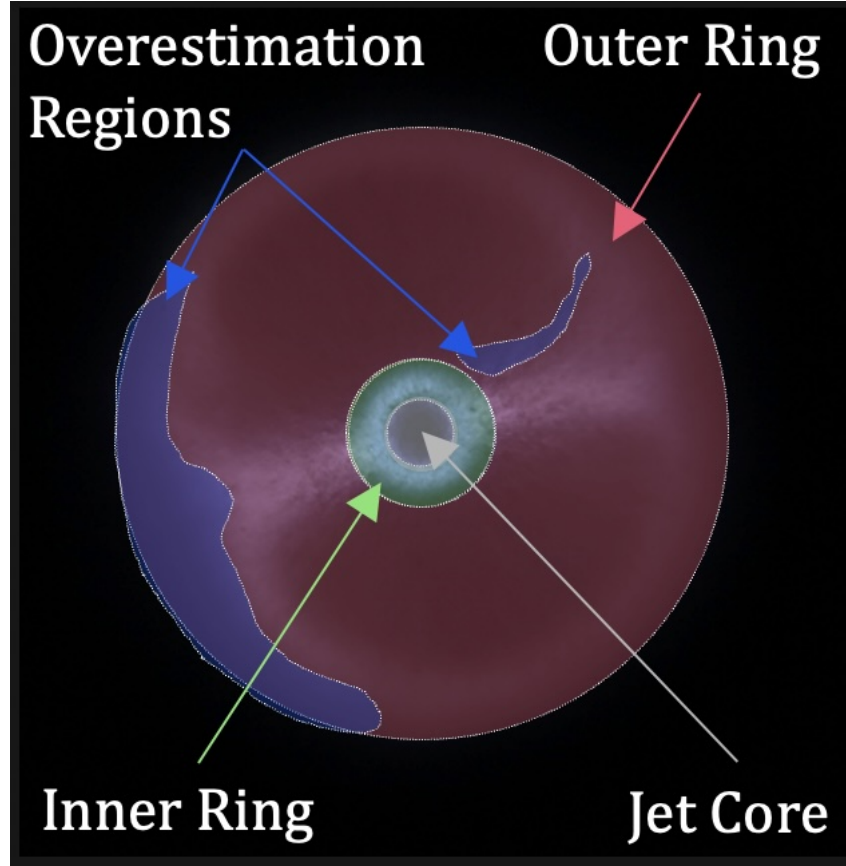


Figure 2.8: Diagram of the plasma regions referred to in the text for analysis. The inner ring, outer ring, primary overestimation regions, and jet core are labeled.

surface roughness. However, given the potential error, especially in the interior ring, this cannot be concluded with certainty without further investigation.

Nevertheless, two distinct plasma phases emerge that cannot be attributed to $\mathcal{R}$ overestimation or small shifts in plasma size. The inner toroidal ring, which appears bright blue in Fig. 2.7(a), exhibits electron temperatures of $T_e \sim 15$ eV, much higher than the ~ 1 eV of the outer diffusive plasma. Although the error in T_e is significant, the red/green bremsstrahlung balance and the elevated T_e clearly indicate a distinct phase in the inner ring. This T_e suggests two key effects: an elevated T_e that produces a blue-shifted bremsstrahlung continuum, and the likely presence of Ar II species at other wavelengths due to argon's ionization energy (15.76 eV). Thus, the blue appearance of the inner ring is likely due to both factors. The transition between the inner high- T_e phase and the outer low- T_e phase occurs over less than $10 \mu\text{m}$, suggesting strong stabilization and tight confinement of the hotter inner toroidal

ring.² In the outer diffusive region, T_e rapidly cools to below 1 eV, consistent with collisional electron cooling and a cascade-type discharge, where the energy of hot electrons is distributed among other ejected electrons.

Electron Density

By considering the population ratios of the ArI transitions at 738 nm, 750 nm, and 763 nm, the mean electron density throughout the plasma can be estimated as $n_e \approx 2 \times 10^{15} \text{ cm}^{-3}$ at $T_g \approx 4500 \text{ K}$ (Zhu, Pu, et al., 2009). Since this is based on total ArI populations, this mean value comprises contributions from both the higher and lower T_e regimes. The complementary 750/738 nm ratio, however, exceeds the Boltzmann thermal limit predicted by the argon-only collisional–radiative model (Zhu and Pu, 2010), indicating additional quenching pathways from impurity species (H_2O , O_2) (Sadeghi et al., 2001) and non-Maxwellian electron-energy distributions. A more robust estimate would require higher-order collisional–radiative models that incorporate molecular-impurity chemistry, non-equilibrium electron-energy distributions, and resonance-line radiation trapping, which is beyond the scope of this chapter.

Similar to T_e , we seek to gain a spatially resolved concept of electron density. To do so, Eq. 2.1 can now be arranged to isolate n_e as

$$n_e = \sqrt{\frac{\lambda m_e}{2n_a^2}} (\kappa T_e)^{3/2} \frac{\epsilon_{\text{ea}}(\lambda_g, T_e)}{\mathcal{I}(\lambda_g, T_e)}. \quad (2.5)$$

Here, λ is taken to be the center wavelength of B_g ($\lambda_g = 507 \text{ nm}$), where ϵ_{ea} has been measured. By normalization, fixed coefficients can be dropped, giving a local relative electron density $n_e/n_{e,\text{max}}$ by the proportionality

$$\frac{n_e}{n_{e,\text{max}}} \propto (T_e)^{3/2} \frac{\epsilon_{\text{ea}}(\lambda_g, T_e)}{\mathcal{I}(\lambda_g, T_e)}. \quad (2.6)$$

Analogous to T_e , this gives a unique 2:1 mapping of $(\epsilon_{\text{ea}}, T_e) \rightarrow n_e/n_{e,\text{max}}$ (see C). Note that in this calculation, the measured intensity, $\mathcal{I}$, is normalized by the approximate plasma thickness, because the images represent a line-of-sight integration of light. The function used to approximate this thickness is included in the supporting

²The sharp spatial boundary is an observation; the stabilizing mechanism—whether hydrodynamic, electromagnetic, or a combination—is addressed in Chapter 5.

information. Furthermore, the plasma edges have been removed from this calculation, as near the border, insufficient plasma thickness information is known. Error estimates, based on those in T_e have been included.

The resulting $n_e/n_{e,\max}$ map, which spans four orders, is shown in Fig. 2.9. The features of n_e closely, but inversely, parallel those from the T_e map. The inner toroidal ring exhibits a calculated $n_e/n_{e,\max}$ ranging from 10^{-3} to 10^{-4} , while the outer ring spans 10^{-1} to 10^0 . As before, the outer edge of the left streamer exhibits edge effects, stemming from the underlying edge effect in T_e . Similarly, we see asymmetry in the streamers reflected in the density.

Depression of n_e in the inner ring supports both that electron-neutral, not electron-ion, bremsstrahlung is the main blue-light emission process and that the radial discharge process is a cascade-type process. As additional electrons are ejected, a cooler plasma phase forms in which the ArI system is pumped and low- T_e ϵ_{ea} occurs. The stark spatial separation in both n_e and T_e between outer and inner-ring phases indicates a unique self-confining behavior that is atypical of such plasmas.³ This intense gradient combined with a stable geometry suggests the possibility of intrinsic magnetization as a stabilization mechanism.

2.4 Conclusion

Optical emissions from a toroidal argon plasma generated through hydrodynamic shear were analyzed to conclude that the main source of visible light is electron-neutral bremsstrahlung radiation. To our knowledge, this is the first confirmation of bremsstrahlung radiation emitted from a plasma created without external sources of voltage or current, but from a purely triboluminescent source. Furthermore, it is a prime example of such, in which bremsstrahlung is the primary optical emitter. Such sources of optical continua are useful as calibration sources for optical systems. The unique feature of this plasma (e.g. having no requirement for current-carrying coils) offers the potential for plasma or RF sources in places where such devices are typically not permitted or detrimental, such as in many medical imaging contexts.

By applying a bremsstrahlung model to images of the red and green light from the plasma, the electron density n_e and temperature T_e were spatially determined. From this, we identify that even within the plasma's well-defined geometry, there are sub-scale structures within the plasma in the form of distinct phases that are

³The analysis of Chapter 5 shows that the azimuthal self-field exerts an outward (loop-expanding) magnetic pressure; the observed spatial confinement may be hydrodynamic rather than electromagnetic in origin.

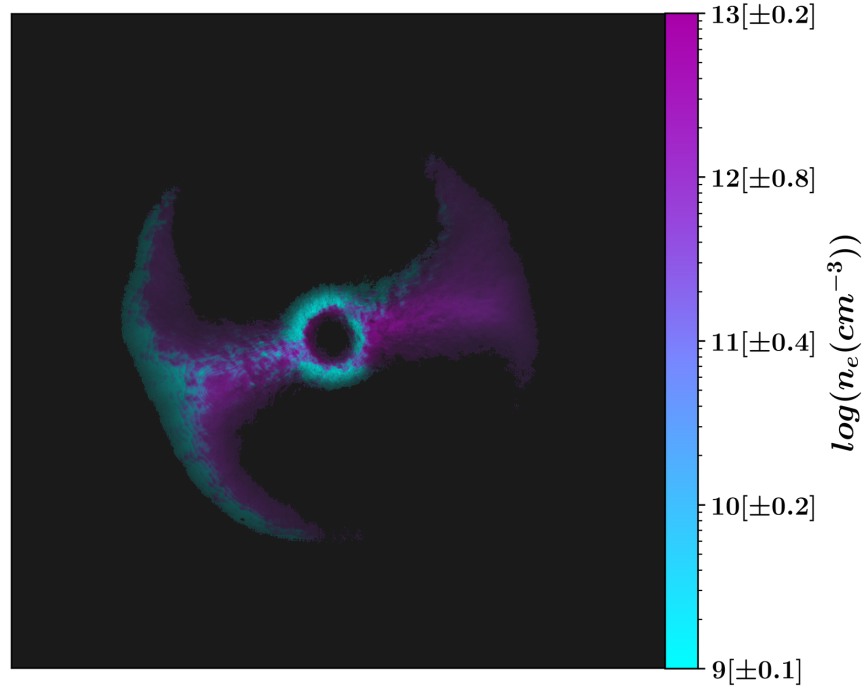


Figure 2.9: Map of log electron density calculated from the normalized original image intensity $I/I_{\max}$ and simulated intensity calculated from B_g and T_e . Error estimates included, based on those in T_e and $\mathcal{I}$.

separated starkly in space. An inner toroidal ring manifests near the cathode as a high T_e /low n_e plasma, which generates a population of ionized argon atoms belonging to Ar II, and bright blue bremsstrahlung radiation. Electrons from this inner ring that are ejected contribute to a cooler bremsstrahlung process that occurs up to a radius of $\sim 500 \mu\text{m}$, along with substantial excitation of neutral argon transitions in the infrared. This secondary process exhibits low T_e ($< 1 \text{ eV}$)/high n_e , which diffuses toward the anode. Neutral argon transitions (Ar I) compose $\sim 99\%$ of the total plasma irradiance. Ionized Ar II transitions compose $\sim 0.1\%$, and electron-neutral bremsstrahlung radiation (ϵ_{ea}) composes $\sim 1\%$ of the total plasma emissivity, corresponding to $\sim 90\%$ of visible light. Thus we ascribe the appearance of the outer plasma to low-energy ($\sim 1 \text{ eV}$) ϵ_{ea} , appearing mostly white, and the appearance of the inner toroidal ring to a combination of higher-energy ($\approx 15 \text{ eV}$)

ϵ_{ea} and Ar II transitions. Notably, the transition between these phases occurs over an extremely short length, about only $10\text{ }\mu\text{m}$, with the inner phase maintaining a highly coherent geometry. We conjecture that the stark boundary between these two phases is indicative of a spontaneous self-confinement mechanism intrinsic to the toroidal plasma that is created.⁴ Future investigations will focus on whether the confinement mechanisms at play are more hydrodynamic in nature, electromagnetic in nature, or a unique interplay between the two.

Data Availability

Image data and processing protocol for these experiments have been made available at Mendoza (2024), to facilitate replication and validation. Data includes raw image and processed image files, along with spectroscopic data. Data available from corresponding author upon reasonable request.

2.5 Summary

In this work, we investigate the anomalous appearance of bright visible light from a toroidal argon plasma generated through extreme hydrodynamic shear (see the Gallery, images ArQ1–ArQ4 and ArL1–ArL4). In ambients of nitrogen or helium, the spectral content is easily correlated to the plasma’s color. In an argon ambient however, infrared spectral lines dominate the spectrum, while the plasma appears light blue, nearly white, to the naked eye. We determine that this luminescence is a visible broadband continuum emitted by electrons scattering off neutral argon atoms, a form of radiation called bremsstrahlung radiation (ϵ_{ea}). Using multispectral imaging, we calculate the temperature, T_e , of bremsstrahlung-producing electrons spatially throughout the plasma, up to the ionization energy of argon atoms 15.76 eV . We further provide a method of calculating electron density, n_e , from the same data. We find that in both the T_e and n_e fields, two distinct phases appear; a small inner ring of high T_e , low n_e , and an outer diffusive region with low T_e , high n_e . The boundary between these disparate phases is extremely small, less than $10\text{ }\mu\text{m}$, indicating an element of self-confinement⁵ intrinsic to this plasma configuration.

⁴See Chapter 5 for a revised analysis showing the magnetic self-pressure is directed outward.

⁵See Chapter 5.

Chapter 3

INTERLUDE

We have established the underlying mechanism for the phenomenon of triboluminescence: interfacial charging in regions of extreme hydrodynamic shear, leading ultimately to an atmospheric-pressure plasma discharge. Comparing the plasma structure generated by a rough substrate against that of a smooth substrate indicates very clearly that the fluid shear is essential to this process and can explain the streamer structure that appears on rough surfaces, or at high jet impingement velocities. In every air plasma seen here, a distinct inner core of the plasma appears which assumes a high degree of continuous rotational symmetry. This core corresponds to the region of elevated electron temperature, as determined from the optical characterization of the emitted bremsstrahlung radiation in the case of the argon plasma. What remains is the question of what mechanism leads to this symmetry, and to the bifurcation of the plasma into these thermodynamically distinct regions. To motivate a series of magnetic sensing experiments, it is worth reviewing some analogous physical scenarios which parallel this geometry closely.

For the remainder of this document, we will generally consider only the cases which are continuously rotationally symmetric (invariant to θ). In such a cylindrical coordinate system (with basis $\{\hat{\mathbf{r}}, \hat{\boldsymbol{\theta}}, \hat{\mathbf{z}}\}$), it will be useful to reference two related vector directions: toroidal and poloidal. Toroidal vectors represent a rotation about $\hat{\mathbf{z}}$ (e.g. along the major radius of a toroid), while poloidal vectors are strictly perpendicular to their toroidal counterparts (e.g. aligned with the minor radius of a toroid). In a standard cylindrical basis, a toroidal vector $\vec{\psi}$ and poloidal vector $\vec{\xi}$ have the components

$$\begin{aligned}\vec{\psi} &= 0 \hat{\mathbf{r}} + \psi_{\theta}(r, z) \hat{\boldsymbol{\theta}} + 0 \hat{\mathbf{z}} && \text{(toroidal vector),} \\ \vec{\xi} &= \xi_r(r, z) \hat{\mathbf{r}} + 0 \hat{\boldsymbol{\theta}} + \xi_z(r, z) \hat{\mathbf{z}} && \text{(poloidal vector).}\end{aligned}$$

Fig. 3.1 illustrates the cylindrical coordinate conventions (black), and the corresponding toroidal ($\hat{\boldsymbol{\psi}}$, blue) and poloidal ($\hat{\boldsymbol{\xi}}$, red) directions.

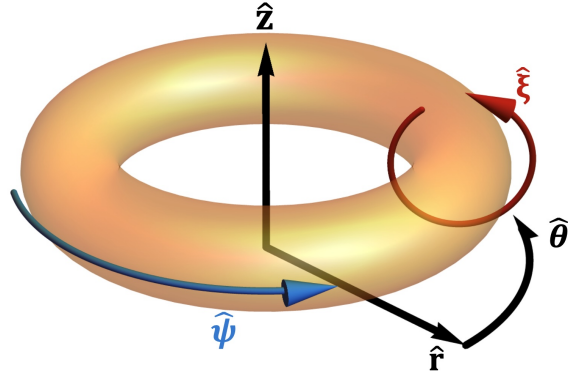


Figure 3.1: Illustration of the cylindrical, toroidal, and poloidal coordinate conventions with an axially oriented toroid as a reference. Unit vectors $\hat{z}$, $\hat{r}$, and $\hat{\theta}$ correspond to the cylindrical coordinate system, while $\vec{\psi}$ (blue) and $\vec{\xi}$ (red) correspond to toroidal and poloidal vectors, respectively. Note that by this convention, $\vec{\psi}$ is parallel to $\hat{\theta}$, and $\vec{\xi}$ always lies in an $r - z$ plane.

A charged particle with finite velocity in a background magnetic field $\vec{\mathbf{B}} = B_z \hat{z}$ follows a toroidal path with fixed radius about the $\hat{z}$ axis, obeying the Lorentz equation

$$\vec{\mathbf{F}} = q \vec{v} \times \vec{\mathbf{B}},$$

with an orbital (Larmor) radius

$$r_L = \frac{m v_{\perp}}{|q| B}.$$

Conversely, a charged particle may travel at fixed velocity in a toroidal magnetic field $\vec{\mathbf{B}} = B_{\theta} \hat{\theta}$, provided it travels along the central axis (such as the current and magnetic field configuration shown in Fig. 3.2(a)). Extending this by projecting the current into a circular loop, and revolving that loop around a central axis (see Fig. 3.2(b)), we see that the associated magnetic field (inside the loops) is toroidal. Analogously, when current is projected along a purely toroidal path, the magnetic field is instead a poloidal field (see Fig. 3.2(c)). An arbitrary superposition of the two produces a helical magnetic field (such as in Fig. 3.2(d)), which is an element of the fundamental field topologies for confining ensembles of charged particles. This most basic configuration illustrates how magnetic fields can confine ionized species, with the underlying toroidal symmetry being favorable.

An additional geometric analogy of interest is the toroidal moment, an independent term of an electromagnetic multipole expansion which can describe the constituent elements of these toroidal structures. The axial toroidal dipole moment is associated

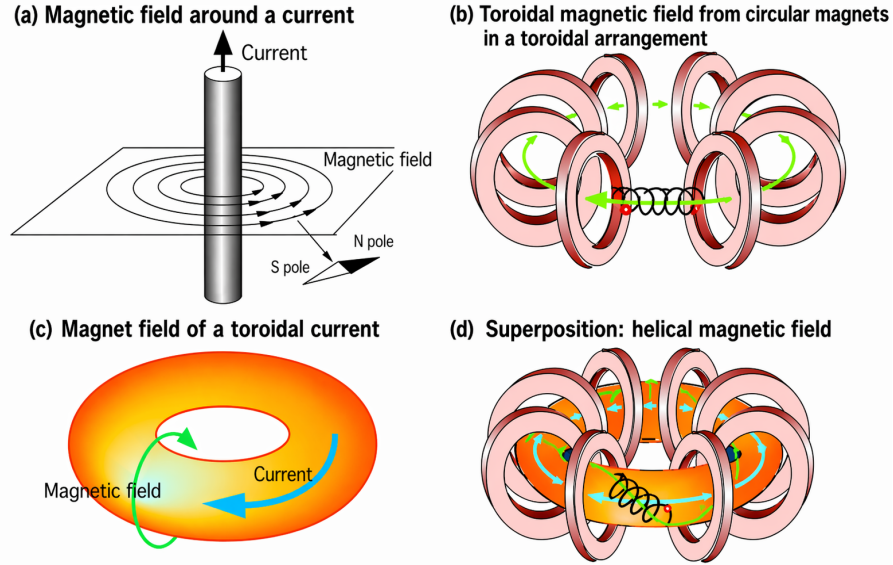


Figure 3.2: (a) Toroidal magnetic field lines circulating around an electric current and a hierarchy of associated toroidal structures. (b) A toroidal magnetic field produced by a series of circular magnetic coils. (c) Magnetic field associated with a toroidal current. (d) Superposition of toroidal and poloidal structures (b) and (c), resulting in a helical field line (green). Adapted with permission from Kikuchi (2010) under CC BY 4.0.

with poloidal currents, and the poloidal toroidal dipole moment is associated with toroidal currents. The phenomena described above are linear superpositions of these fields, intended to confine plasma into the shape of a torus. We, however, would like to bring in the idea of a toroidal dipole superposition with a parallel electric polarization. In particular, an electric dipole and a toroidal dipole with aligned moments can produce an anapole-like structure, in which case the continuum symmetry of these fields can exhibit full cancellation in the far field, resulting in a non-radiative multipole.

Recall from Chapter 1 (Fig. 1.1) the discharge model, which explicitly identifies the electric current structure as poloidal in nature. Consequently, we would expect an associated toroidal (azimuthal) magnetic field to arise. As shown in Chapter 5, the B_θ^2 magnetic pressure of such a field acts *outward*, tending to expand the current loop rather than confine it. A purely toroidal field cannot confine charged species; however, the question of whether it influences the plasma morphology at all—for instance, by increasing transit time through gyration, or by coupling to the impinging-jet hydrodynamics—remains open.

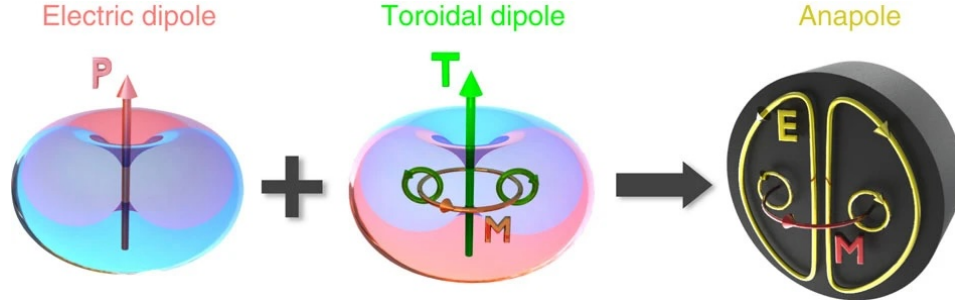


Figure 3.3: The structure of a so-called anapole state, comprising an electric dipole (left) and a toroidal dipole (middle), aligned and superposed (right). When the magnitude of each is equal and perfectly out of phase, the anapole is non-radiative in the far field. Reproduced with permission from Miroshnichenko et al. (2015) under CC BY 4.0.

From Chapter 1 and Appendix B, we calculated electron pressures of approximately 10–68 kPa and ~ 390 Pa for the plasmas established in nitrogen and argon, respectively. The magnetic field that would be *required to balance* the upper end of each electron pressure—i.e. with magnetic pressure $B^2/(2\mu_0)$ set equal to $n_e k_B T_e$ —is up to approximately 290 mT and 31 μ T, respectively. This is a hypothetical equivalence used to set a scale; the electron pressure does not itself produce a magnetic field. Bearing in mind that spatial averaging likely exaggerates the effective electron temperature, we can nevertheless ask whether a self-induced magnetic forcing of this magnitude is feasible at all.

From Appendix B, a discharge current density of $J \approx 8 \text{ mA } \mu\text{m}^{-2}$ (air plasma) was estimated from spectroscopic data. We stress that the resulting current is *highly uncertain*: it rests on a spectroscopically inferred current density together with an assumed plasma thickness $\delta \approx 10 \mu\text{m}$ and an assumed core radius $R \approx 100 \mu\text{m}$. Under these assumptions the *instantaneous peak* discharge current is

$$I \approx J \delta (2\pi R) \approx \left(8 \frac{\text{mA}}{\mu\text{m}^2} \right) \underbrace{(10 \mu\text{m})}_{\text{plasma thickness}} \underbrace{(2\pi \cdot 100 \mu\text{m})}_{\text{circumference}} \approx 50 \text{ A}.$$

This peak applies only to the active discharge of the air/nitrogen plasma; because the discharge is pulsed, its time-averaged current is far smaller (quantified in Chapter 5). By Ampère’s law, the corresponding azimuthal field at the core radius is $O(100 \text{ mT})$,

$$B_\theta \approx \frac{\mu_0 I}{2\pi r} \approx 100 \text{ mT},$$

of the same order as the field that would balance the electron pressure, estimated above.

Signature of an Enclosed Poloidal Current

For a closed poloidal current circuit with cylindrical symmetry, the azimuthal field is fixed by the *enclosed* current alone. By Ampère’s law,

$$B_\theta = \frac{\mu_0 I}{2\pi r},$$

where I is the net current enclosed by a circle of radius r ; the field vanishes in any region that encloses no net current. The succeeding chapter, however, measures the field *outside* the enclosed current—below the target plate—where a perfectly axisymmetric poloidal current would produce no field. A nonzero measurement there therefore probes the *non-axisymmetric* structure of the current, rather than directly confirming the idealized axisymmetric enclosed-current field.

Chapter 4

IN SEARCH OF A MAGNETIC SIGNATURE: DESIGN OF A WALL-INTEGRATED MAGNETOMETER FOR NEAR-FIELD SENSING

Manuscript in preparation for submission to Nature Communications.

4.1 Background

It is well known that with appropriate control of external electric and magnetic fields, plasma can be manipulated and confined. Hallmark examples include the toroidal tokamak (Taylor, 1997), pinch plasmas (e.g. z-pinch (Lieberman et al., 1999) or θ -pinch (Loisch et al., 2014)), and spheromaks (Jarboe, 1994), among others. In the aforementioned structures, electromagnetic coils are driven with the goal of counteracting the intrinsic pressure of the plasma. In each of these structures, axisymmetry is leveraged in some way to achieve stability against plasma heating. In Chapter 2, we showed that a distinct thermal phase exists in our class of atmospheric toroidal plasmas which demonstrates an axisymmetry that is unique among high-pressure natural plasmas. Previously, we showed that these plasmas have a coherent DC polarization; a negative electrode (cathode) corresponds to the impinging neck of the jet, and the positive electrode (anode) to the downstream flow where the fluid layer thins. The fundamental mode of this plasma is a toroid with axial symmetry (see the Gallery, images AQ8, AL3, AL7, AL9, ArQ2, ArQ4, ArL1–ArL3, HeQ1, and HeL1), which has yet to be explained via experiments. We hypothesize that a self-generated magnetic field accompanies this current-carrying plasma and seek to determine its structure, magnitude, and role—if any—in the plasma’s morphology. Prior measurement attempts with standard Hall effect magnetometers have yielded null measurements, and were found to substantially modify the plasma shape when in close proximity, offering a current-sink path. To measure this system non-invasively, we turn to a magnetic resonance technique based on optical detection of nitrogen-vacancy (NV^- center) defects in diamond.

Magnetic Sensing Challenges—Scale and Sensitivity

Measuring fields in the plasma’s near-field poses two distinct challenges. First, given the plasma length scale, $\sim 100\text{ }\mu\text{m}$ – $500\text{ }\mu\text{m}$, the total current associated

with the discharge is expected to be small ($O(10\text{ mA})$ on average; see Chapter 5), distributed as a toroidal moment composed of axial and poloidal current elements with a characteristic scale of $\sim 100\text{--}500\text{ }\mu\text{m}$. Correspondingly, any unique field structure that might be sensed externally should exhibit a similar spatial extent. Thus, an approach with good sensitivity is needed. The second challenge is that the measurable field strength scales strongly (with negative power) with distance from the source. Dipole fields of infinitesimal size, including magnetic dipole fields, decay as $1/r^3$. Because our system is nearly axisymmetric, a perfectly symmetric poloidal current would produce no external azimuthal field (Chapter 5); any externally measurable signature therefore arises from departures from perfect axisymmetry, which manifest as higher-order multipole terms that decay even more steeply with distance. Qualitatively, configurations of 2-, 4-, and 8-fold discrete symmetry and the continuum limit of this symmetry are shown in Fig. 4.1(a). The analytic fields for each case are shown in Fig. 4.1(b), illustrating that the field strength falloff scales strongly with pole multiplicity. Thus, successful measurement of the hypothesized fields depends intimately on both measurement sensitivity and proximity.

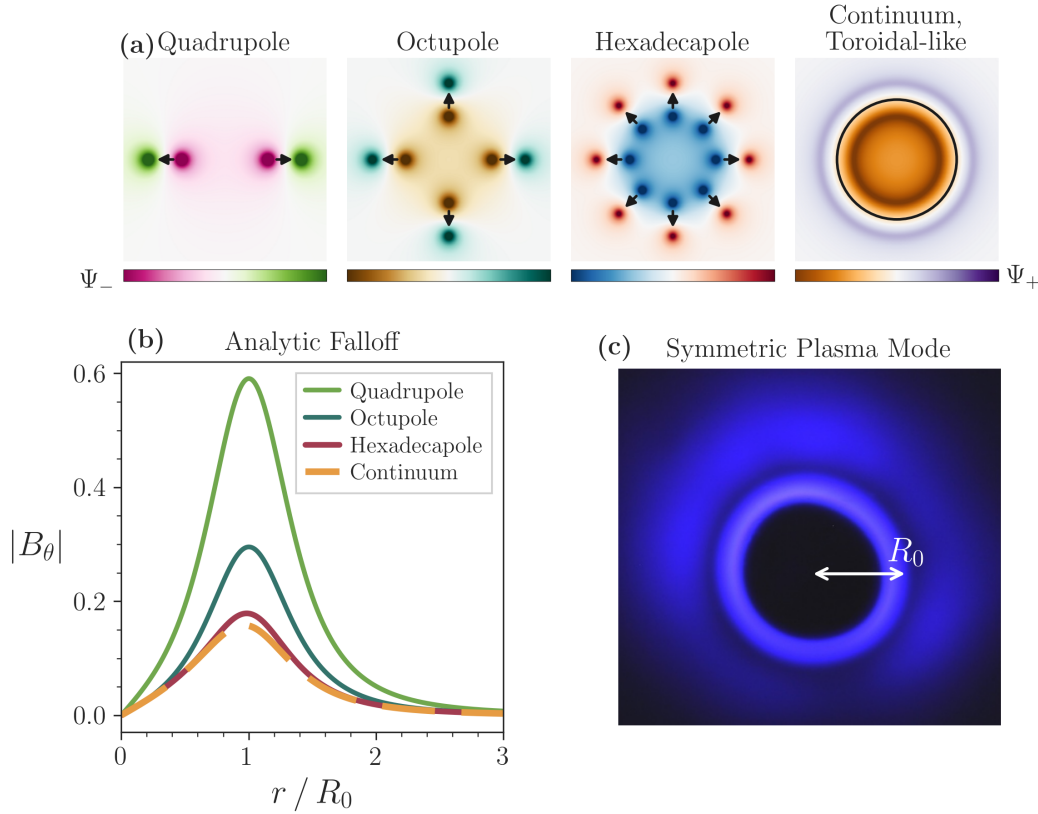


Figure 4.1: Multipole configurations and field decay. (a) Multipole configurations of a toroidal magnetic field, B_θ , with increasing (left to right) N -fold rotational symmetry, and the continuum limit of the axisymmetric expansion (right, analogous to a toroidal moment). (b) Analytic field decay curves for corresponding multipoles, showing decay rate growing with order, most aggressive for the continuum case. (c) Configuration of the fully symmetric, stable plasma, structurally analogous to the continuum case. These distribution properties apply both to the electric field from electric multipoles and to the transverse magnetic field from infinitesimal current elements (indicated by black arrows, $\rightarrow$).

To address both challenges, we chose the magnetically sensitive nitrogen vacancy (NV^-) defect in diamond as the fundamental sensing unit. By embedding a monocrystalline diamond into the wall of our shear flow, the magnetism-dependent fluorescence of the NV^- centers can be remotely read without flow interference. With this approach, the sensing bodies could be placed within $\sim 75 \mu\text{m}$ of the water-solid interface, and achieve a practical sensitivity of $120 \text{ nT Hz}^{-1/2}$. Fig. 4.2 shows schematically the fundamental components of this sensing approach, including the water jet configuration, embedded magnetometer, and optics.

While existing magnetometer technologies easily boast impressive sensitivities—e.g. SQUID at $O(100 \text{ aT Hz}^{-1/2})$ (Drung et al., 2007); SERF/vapor cell magnetometers at $O(1 \text{ fT Hz}^{-1/2})$ (Allred et al., 2002)—that exceed ours (with reports of down to $9 \text{ pT Hz}^{-1/2}$), the NV center maintains unique technical advantages: nm-scale sensing units ($\sim 4 \text{ \AA}$ unit-cell, $\sim 4 \text{ nm}$ achievable particle size (W. H. Bragg and W. L. Bragg, 1913; Hudak and Stroud, 2023)) and optical readout that allows for measurement without interference with the system¹. Contrasted against SERF or SQUID magnetometers, which require at minimum thermal control (heating/liquid He or N₂), NV[−] centers are uniquely positioned for this task of μm -scale magnetic sensing.

¹Within engineering constraints of incorporating these sensors. See also (Wickenbrock et al., 2016) for a discussion of a microwave-free version of the technique.

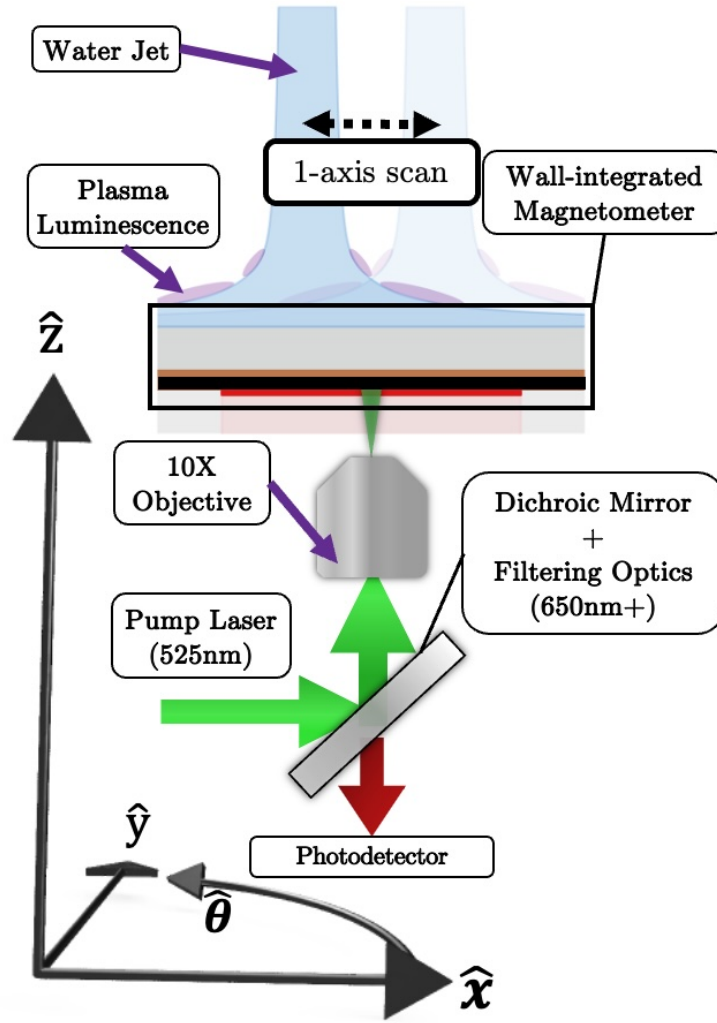


Figure 4.2: Paradigm for the wall-integrated magnetometer for near-field flow sensing, showing the flow configuration for our setup as the water jet which impinges on the flow wall with integrated sensing layers. A simplified schematic of the sensing optics is shown probing the diamond layer (red), used for remote sensing. Lab frame coordinate axes are shown ($\hat{x}$, $\hat{y}$, $\hat{z}$, $\hat{\theta}$).

At present, various interests and embodiments of this NV^- center-based technique include large-scale flow pattern detection (M. K. Fu and Dabiri, 2023), (magneto)cardiograms (Yu et al., 2024; Zheng et al., 2021), ultra-slow flow sensing (Fernández-Acebal and Plenio, 2018), mobile sensors of flow (Andrich et al., 2014), or detecting ion transport *in situ* in batteries (Hollendonner et al., 2023). Nowodzinski et al. (2015) demonstrate an approach to resolving μm -scale current structures in integrated circuits, which they dub *Magnetic Current Imaging (MCI)*.

This work demonstrates the first such approach, to our knowledge, that is applicable to multiphase (liquid, solid, gas, plasma) extreme flow conditions ($Re \sim 20000$ – 50000). We apply this technique to both calibration scenarios with fixed coils, and to the scenario of an axisymmetric air plasma generated through extreme hydrodynamic shear (Fig. 4.1c) and a fundamental measurement of the magnetic-current component of the shear flow of pure water.

4.2 Methods

Magnetic Resonance with NV^- Centers

Fluorescent nitrogen-vacancy (NV^-) centers in diamond are an extremely well-characterized host of quantized energy levels which have been studied for decades for use in both electromagnetic and mechanical (temperature, strain) sensing (Boretti et al., 2019; Loubser and Wyk, 1978). Photodetection of the fluorescence of single NV^- defects has been achieved, allowing for true nano-scale sensing of these fields.

To enable the application of this sensing to microfluidic flows, we design and demonstrate an NV^- -center based magnetometer configuration that can be integrated into the wall of fluid systems. This system was primarily designed to enable magnetic sensing of micrometer and microtesla-scale magnetic moments resulting from high-speed microjets of water, but can easily be adapted for sensing of temperature or electric fields in the near-field of fluid systems. This configuration comprises an epi-fluorescence microscope which optically initializes an NV^- center ensemble in bulk diamond positioned within a distance of $75\ \mu\text{m}$ from the fluid, with integrated optical, electrical and mechanical shielding. This extreme proximity allows better detection of the generated moments, expected to be on the nanotesla scale, and allows for some spatial resolution of NV^- states without disrupting the fluid flow. With this system, we achieve a shot-noise-limited sensitivity of $9\ \text{nT Hz}^{-1/2}$ and a technical-noise-limited sensitivity of $120\ \text{nT Hz}^{-1/2}$.

Fundamentally, NV^- center sensing is a pump-probe based spectroscopy technique, which can be operated as a remote sensing technique. A schematic showing the optical pump-probe configuration and the integrated sensor relative to our flow system of interest is shown in Fig. 4.2. Here, we employ a continuous-wave (CW) method whereby CW-laser-induced photoluminescence (PL) is measured while a frequency-swept CW-microwave (MW) field is applied to identify resonant frequencies. For thorough characterization of the NV^- center's resonance spectra and dynamic-pulsed techniques, the reader is directed to resources such as (Boretti et al.,

2019; Loubser and Wyk, 1978), or Schirhagl et al. (2014), Degen, Reinhard, and Cappellaro (2017), and J. M. Barry et al. (2016), all of which provide a deep review of certain applications and approaches for enhancing the technique's sensitivity.

This detection technique, through measuring the resonance spectrum of the trapped electron associated with NV^- centers, provides sensitivity to various fields represented through the NV^- center's eigenstates furnished by its Hamiltonian. The spin Hamiltonian gives the NV^- center's energy eigenstates as a function of local fields and is cited by Schirhagl et al. (2014) as

$$\begin{aligned} \frac{\mathcal{H}}{\hbar} = & \underbrace{\epsilon_z E_z \left(S_z^2 - \frac{2}{3} \right) + \epsilon_{xy} \left\{ E_x (S_x S_y + S_y S_x) + E_y (S_x^2 + S_y^2) \right\}}_{\text{Electric}} \\ & + \underbrace{D \left(S_z^2 - \frac{2}{3} \right)}_{\text{ZFS}} + \underbrace{\gamma \vec{\mathbf{B}} \cdot \vec{\mathbf{S}}}_{\text{Magnetic}}, \end{aligned} \quad (4.1)$$

where ϵ_{ij} are strain tensor elements, E_i and B_i are electric and magnetic vector elements, D is the zero-field splitting parameter (~ 2.879 GHz), and S_i are the spin vectors. Under a magnetostatic field, the energy states of an NV^- center exhibit resonance frequencies given to first order by the following expression.

$$f_{\pm} = D \pm \gamma B = 2.879 \text{ GHz} \pm 28 \text{ MHz mT}^{-1} \cdot B \quad (4.2)$$

indicating that the presence of a nonzero magnetic field generates a pair of resonance frequencies belonging to the ± 1 spin state separated by a factor proportional to the component B_z , where z is the NV^- -center axis. The vector sensing regime is illustrated schematically in Fig. 4.3, showing the four diamond lattice basis vectors (a), the pump-probe energy level diagram (b), and the spectral characteristics coupled to the magnetic field (c).

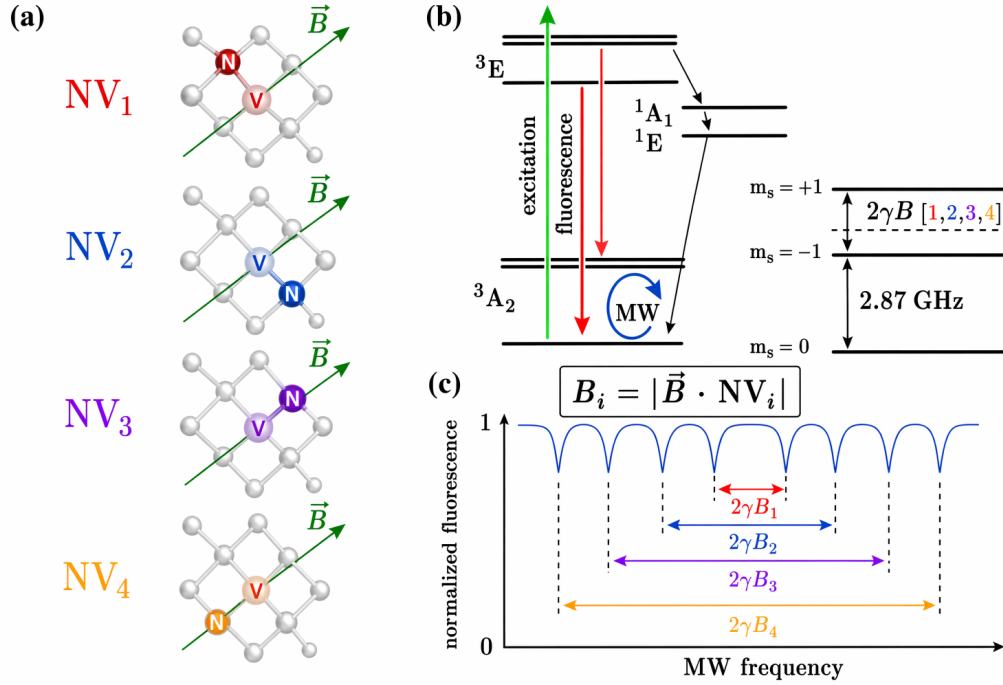


Figure 4.3: Diagram illustrating the NV^- center magnetic sensing scheme. (a) Possible NV^- center orientations with respect to a constant background magnetic field vector $\vec{B}$. (b) Schematic of the excitation, radiative decay, and non-radiative decay energy levels, which are spin-state-dependent ($m_s = -1, 0, +1$). (c) Normalized fluorescence spectrum (blue) with magnetic field components labeled in the $\vec{NV}_i$ basis, where each component is given to lowest order by $B_i = |\vec{B} \cdot \vec{NV}_i|$. Adapted with permission from Homrighausen, Horsthemke, et al. (2023) under CC BY 4.0.

The presence of a transverse electric field similarly generates a resonance split with coupling coefficient $0.17 \text{ Hz V}^{-1} \text{ m}$, which is also captured by the Hamiltonian. By applying a strong fixed bias magnetic field with known amplitude and electrical shielding, we demonstrate measurement of purely a magnetostatic field in the near field of this extreme wall shear flow.

4.3 System Design

In this system, CW optically detected magnetic resonance (CW-ODMR) is carried out using a fluorescence microscopy configuration in which an excitation beam, directed through microscope optics, illuminates the fluorescent sample and is subsequently read out. The emitted fluorescence (red) is filtered out from the incident beam (green) through several optical filters. A functional block diagram of the signal pathway and communication between signal, power control, and output data is

shown in Fig. 4.4. Technical parameters of its constituent subsystems are described below.

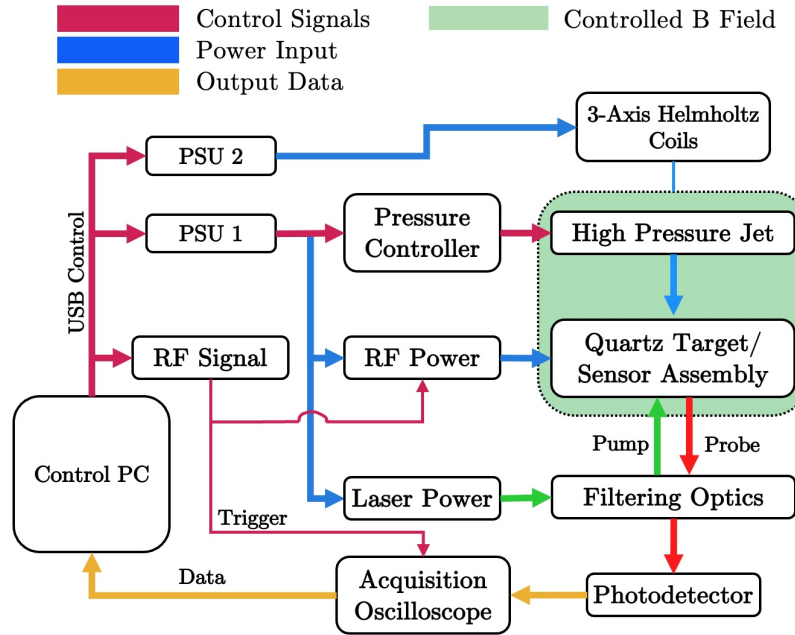


Figure 4.4: Functional block diagram showing the communication structure of this experiment. Signal types are labeled according to the legend. Engineered optical laser beam paths are shown with green ($\uparrow$) and red ($\downarrow$) arrows for the pump and probe signals, respectively. The sensor assembly and associated test section elements are all contained within the region of controlled B field.

Laser Excitation

A CW 1.2 W, 525 nm fiber laser (LaserTack FL-525-1200) provides the excitation beam which initializes the NV^- center spin state and produces fluorescence for readout. The incident beam is first collimated, then manipulated through refocusing optics and reflected (Thorlabs DMLP567R dichroic mirror) to the back focal plane of the objective (Nikon Plan 10X, NA 0.3). The focus on this plane results in wide-field illumination exiting the objective (Fig. 4.3). The beam volume is directed to the NV^- -doped diamond sample (Diatope, custom fabrication: 4 mm $\times$ 4 mm, 5 μ m NV^- layer). The laser is operated at about 20% power to optimize the sensitivity of the photodetector, resulting in a beam of about 240 mW.

RF Modulation

A vector signal generator (Signalhound VSG60, bandwidth 40 MHz) provides IQ based frequency modulation of the microwave (MW) field. The generator was set to -5 dBm output which is then amplified via a MiniCircuits $+47$ dBm amplifier (ZHL-16W-43+). The frequency is positively swept with a 20–40 ms period in a continuous positive sweep through a range of ~ 2.86 – 2.90 GHz.

Photodetection

The photoluminescence from the resulting resonance spectrum is viewed through the same $10\times$ objective and passed through two stacked edgepass filters (650 nm longpass, Thorlabs FELH0650) for high isolation between the incident and emergent beams. Images are taken with a QHY 533m (monochrome) or ZWO ASI533MC camera (color). For rapid, single-point acquisition, the signal from a 100 kHz amplified avalanche photodetector (Thorlabs APD440A2) is passed through a 1 kHz low-pass filter (EF110) into an oscilloscope (Tektronix MDO3024, 2.5 GS s^{-1} , or Digilent ADP2230) in AC coupling mode. A mechanical iris in front of the photodiode allows narrowing of the collection window to the desired spot size.

A table of OEM equipment used in this configuration is provided in Table 4.1.

Table 4.1: OEM equipment used in the epi-fluorescence magnetometry setup.

Subsystem	Item	MFG	P/N	Notes
Laser	Fiber Laser	LaserTack GmbH	FL-525-1200	525 nm, 1.2 W
Optics	Objective	Nikon, Inc.	Plan 10X (NA 0.3)	567 nm cut-on
Optics	Dichroic Mirror	Thorlabs	DMLP567R	
Optics	Cage Cube	Thorlabs	CM1-DCH	
Optics	Iris	Thorlabs	SM1D12SZ	
Optics	Lens Adjuster	Thorlabs	SM1ZM	
Optics	Quick Adapter	Thorlabs	SM1QA	
Sensor	NV ⁻ Doped Diamond	Diatope	DT145	5 μ m Layer
Camera	Imaging Camera	QHY	533m	Monochrome
Camera	Imaging Camera	ZWO	ASI533MC	Color
Detection	Avalanche Photodetector	Thorlabs	APD440A2	100 kHz
RF	Vector Signal Generator	Signalhound	VSG60A	6 GHz, 40 MHz BW
RF	RF Amplifier	MiniCircuits	ZHL-16W-43+	0.8–4.2 GHz, +47 dBm
RF	RF Antenna	JLCPCB	Custom	
Acquisition	Oscilloscope	Digilent	ADP2230	125 MS s ⁻¹
Power	Current Source (DC PSU)	Uni-Trend US	UDP3305S-E	10 mV/1 mA res.

Custom Sensor Assembly

Layered Sensor Elements

The core of this configuration is the layered sensor assembly, which allows field detection with extreme proximity to our flow or structures of interest, comprising the NV⁻ diamond, antenna, optical window, and flow wall (jet target). Fig. 4.5 depicts the ordered sensor layers: Optical window (1 mm thickness, ITO-coated borosilicate) → flex PCB antenna (custom fab, design adapted from Ma et al. (2019)) → NV⁻-doped diamond → 10 μ m copper foil → 50 μ m quartz wall → Flow region.

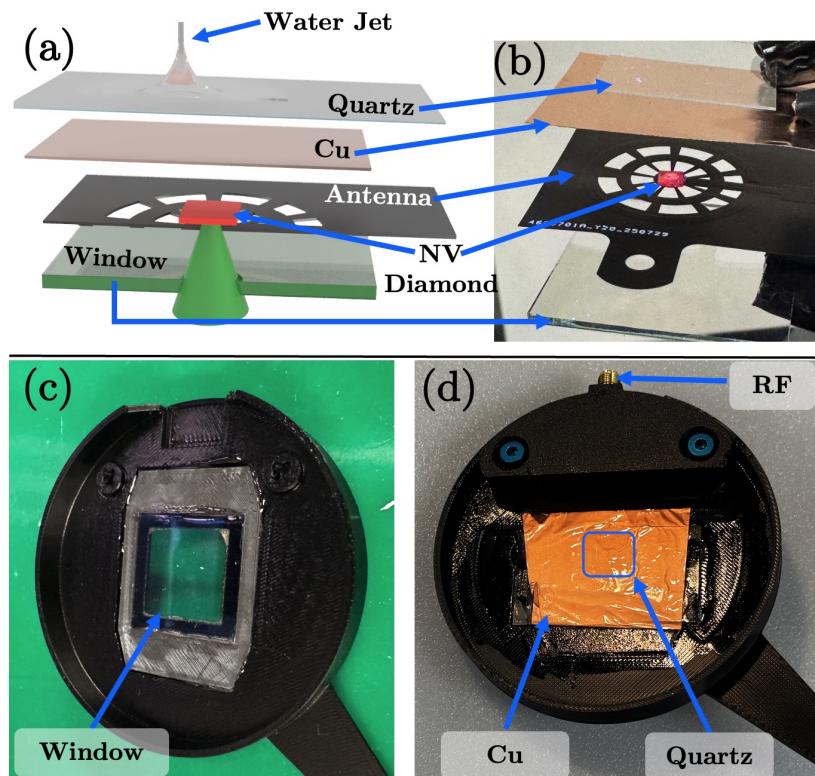


Figure 4.5: Exploded view CAD render (a) and photograph (b) of the sensor assembly layers, shown separated for clarity; RF connector is not shown. 3D-printed mounting plate for the optical window (c), and the final mounted configuration (d) with mounted RF connector shown.

Assembly

The thin quartz film (50 μ m) is placed on a quartz plate (2 mm thickness) which has been coated with epoxy mold release (MG Chemicals epoxy mold release), facilitating the release and holding of the delicate film passively in place. A sheet

of 10 μm copper foil is cut to $\sim 50\text{ mm} \times 50\text{ mm}$ and placed, along with the quartz and NOA61 adhesive in an oven at 50°C for 1 hr to reduce the adhesive's viscosity. A drop of adhesive ($\sim 25\text{ }\mu\text{L}$) is placed on the quartz film and sandwiched between the film and the copper foil, using a rubber stamp to distribute pressure. In the pressed configuration, a handheld UV gun is used to cure the adhesive in place. On this quartz-foil laminate, additional adhesive (about $50\text{ }\mu\text{L}$) is placed on the copper side, followed by the diamond and then the flex PCB antenna such that the antenna's viewing window is fully aligned with the diamond sample. Fig. 4.5 illustrates the relative positioning of these elements. Additional adhesive is added to fully cover the diamond and antenna (about $100\text{ }\mu\text{L}$). A strip of copper can be folded back, followed by covering the whole assembly with the ITO-coated glass. This provides electrical contact between the glass and the copper foil, wrapping the diamond in a conductive shield. UV curing is then applied from the interrogation side to set the antenna and diamond in place. Finally, an overnight cure with low UV power ensures maximal curing of the adhesive. The sensor layering order, material specifications, source, and notes are given in Table 4.2.

Table 4.2: Assembly order and structure of the layered NV^- -center based magnetic sensor.

Layer	Component	Material / Item	MFG	P/N	Notes
<i>Sensor Stack (top to bottom)</i>					
1	Optical Window	ITO-Coated Glass, $30 \times 30\text{ mm}$	Generic	NA	ITO faces sensor; contact to Cu shield
2	Sensing Crystal	NV^- Doped Diamond	Diatope	DT145	$5\text{ }\mu\text{m}$ doped layer
3	Bonding Agent	NOA81 UV Optical Resin	Norland Products	NOA81	Fast cure, low fluorescence
4	Antenna	Custom PCB Antenna	JLCPCB	Custom*	Adapted from Ma et al. (2019)
5	Shielding Foil	$10\text{ }\mu\text{m}$ Copper Foil	JIMA Copper	NA	
6	Thin Film	$50\text{ }\mu\text{m}$ Quartz	PMO [†]	PSQB-3R8552	
<i>Mechanical Support</i>					
	Sensor Housing	3D-Printed Mount (PETG)	In-House	Custom	Non-magnetic
	Translation Stage	2-Axis Translation Stage	In-House	Custom	Non-magnetic

[†]Precision Micro Optics

*See Appendix E

A quartz film of $50\text{ }\mu\text{m}$ combined with a $10\text{ }\mu\text{m}$ copper foil and a thinned layer of adhesive ($10\text{--}20\text{ }\mu\text{m}$) positions the NV^- layer in the diamond within $70\text{--}80\text{ }\mu\text{m}$ of the quartz-water interface. The copper and ITO coating provide an electrical

shield around the diamond, at minimum restricting the plane of the copper foil to a fixed potential and balancing the potential on both sides of the diamond via the ITO coating.

Experimental Setup and Design

Magnetically Sensitive Regime

In the CW mode (laser and MW power), only DC magnetic field components are sensed. The sensitivity to DC magnetic fields over electric fields is increased by applying a fixed bias field via Helmholtz coils with constant current. Since the NV^- sensing technique is a vector measurement but lacks sign information, the known sign of this offset field allows us to retain sign information via computing offsets. In this regime, where the total applied field is $|B| \sim 2 \text{ mT}$, the resonance frequencies for an NV^- lattice vector $\hat{\mathbf{N}}\mathbf{V}_i$ are given to second order by Homrighausen, Hoffmann, et al. (2024) as

$$f_i^\pm = D + \frac{\gamma^2 B_\perp^2}{D} \pm \sqrt{\gamma^2 B_\parallel^2 + E_{\perp,i}^2 + \frac{\gamma^4 B_\perp^4}{4D^2}} \quad (4.3)$$

The possible NV^- lattice unit vectors, which locally ascribe the meaning of $\perp$ and $\parallel$ of the applied field, are given by:

$$\hat{\mathbf{N}}\mathbf{V}_j \in \frac{1}{\sqrt{3}} \left\{ \begin{pmatrix} 1 \\ 1 \\ 1 \end{pmatrix}, \begin{pmatrix} 1 \\ -1 \\ -1 \end{pmatrix}, \begin{pmatrix} -1 \\ 1 \\ -1 \end{pmatrix}, \begin{pmatrix} -1 \\ -1 \\ 1 \end{pmatrix} \right\}$$

in the diamond frame. The parallel and perpendicular projections of the applied fields onto each NV^- axis are

$$\vec{\mathbf{B}}_{\parallel,i} = (\vec{\mathbf{B}} \cdot \hat{\mathbf{N}}\mathbf{V}_i) \hat{\mathbf{N}}\mathbf{V}_i \quad (4.4)$$

$$\vec{\mathbf{B}}_{\perp,i} = \vec{\mathbf{B}} - \vec{\mathbf{B}}_{\parallel,i} \quad (4.5)$$

$$\vec{\mathbf{E}}_{\perp,i} = \vec{\mathbf{E}} - (\vec{\mathbf{E}} \cdot \hat{\mathbf{N}}\mathbf{V}_i) \hat{\mathbf{N}}\mathbf{V}_i \quad (4.6)$$

$E_{\perp,i}$ is the transverse electric field projection on $\hat{\mathbf{N}}\mathbf{V}_i$ (scaled by $0.17 \text{ Hz V}^{-1} \text{ m}$).

As NV^- center energy is sensitive to both electric and magnetic components of the electromagnetic field, we must ensure that our measurements can decouple the effect of both fields. Based on the Hamiltonian (Eq. 4.1), we see that on a single NV^- center $B_\parallel$ induces Zeeman splitting, while electric fields have primarily a

transverse coupling coefficient [$0.17 \text{ Hz V}^{-1} \text{ m}$], but a weak longitudinal coupling [$3.5 \times 10^{-3} \text{ Hz V}^{-1} \text{ m}$] which shifts, but does not split the resonance peaks. Sensitivity to $\vec{\mathbf{B}}$ is shown by considering the quantity $(\Delta f_i)^2$ from Eq. 4.3. Under the assumption $E_z \gg E_x, E_y$ made possible both by electrical shielding and the system geometry, the perpendicular $\vec{E}$ projection for each NV^- axis is degenerate:

$$E_{\perp,i}^2 = \vec{\mathbf{E}}^2 - (\vec{\mathbf{E}} \cdot \hat{\mathbf{N}}\mathbf{V}_i)^2 \approx \frac{2}{3} E_z^2.$$

So the total relation is

$$(\Delta f_i)^2 = \gamma^2 B_{\parallel,i}^2 + E_{\perp,i}^2 + \frac{\gamma^4 B_{\perp,i}^4}{4D^2}, \quad i = 1, \dots, 4 \quad (4.7)$$

With the electric field restricted by electrostatic shielding ($|E_x| \approx |E_y| \ll |E_z|$) to $\vec{\mathbf{E}} \approx (0, 0, E_z)$, substituting the explicit projections from Eqs. 4.4–4.6 yields

$$(\Delta f_i)^2 = \gamma^2 (\vec{\mathbf{B}} \cdot \hat{\mathbf{N}}\mathbf{V}_i)^2 + \frac{2}{3} E_z^2 + \frac{\gamma^4}{4D^2} (B^2 - (\vec{\mathbf{B}} \cdot \hat{\mathbf{N}}\mathbf{V}_i)^2)^2, \quad i = 1, \dots, 4 \quad (4.8)$$

with the unknown vector of variables

$$\vec{\xi} = (B_x, B_y, B_z, E_z). \quad (4.9)$$

Supposing $E_z \approx 0$, uniqueness is guaranteed if

$$\text{rank}(J_{ij}) = \text{rank} \left(\frac{\partial (\Delta f_i)^2}{\partial \xi_j} \bigg|_{\vec{\mathbf{B}}=\vec{\mathbf{B}}_0} \right) = 3$$

At $E_z = 0$, $J_{i4} = \vec{0}$, so $J \in \mathbb{R}^{4 \times 3}$. At our given background field in this study ($\vec{\mathbf{B}}_0 = (-418, 782, -2283) \mu\text{T}$), $\text{rank}(J) = 3$. Therefore, for a given $(\Delta f_i)^2$, (B_x, B_y, B_z) are uniquely determined. Instead supposing that $E_z \neq 0$, with unknowns $\vec{\xi} = (B_x, B_y, B_z, E_z)$, the system is guaranteed a unique solution provided

$$\det \left(\frac{\partial (\Delta f_i)^2}{\partial \xi_j} \bigg|_{\vec{\mathbf{B}}=\vec{\mathbf{B}}_0} \right) \neq 0 \quad (4.10)$$

which is known to be satisfied for any choice of $\vec{\mathbf{B}}_0$ with non-degenerate elements (up to a sign). Substituting $\gamma = 28.025 \text{ MHz mT}^{-1}$ and $D_{\text{gs}} = 2.879 \text{ GHz}$ gives $\det(\partial(\Delta f_i)^2/\partial \xi_j|_{\vec{\mathbf{B}}_0}) \approx 4.5 \neq 0$. Hence, with knowledge of $(\Delta f_i)^2$, (B_x, B_y, B_z, E_z) are uniquely determined.

Jet impingement

For testing jet impingement, a jet orifice diameter of $125\ \mu\text{m}$ was chosen to reduce substrate damage. Driving pressures of 25–75 psi were used, corresponding to incident flow velocities of approximately $115\text{--}150\ \text{m s}^{-1}$. For each case, a piezo stage mounted to the jet manifold was used to translate the jet over an approximate domain of -350 to $+350\ \mu\text{m}$, ensuring that the full span of the jet, and the entire cathode region of the plasma, would be scanned. The full experimental configuration is shown in Fig. 4.6.

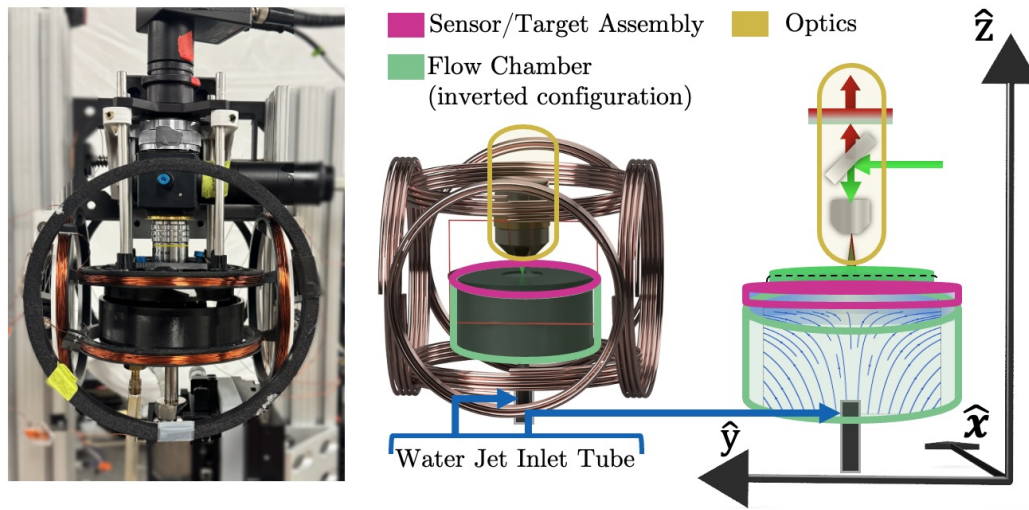


Figure 4.6: Render (a), image (b), and illustration (c) of the experimental setup in the inverted jet configuration used here. The sensor and target assembly position is shown with the optical access window from the top, the flow chamber with inserted water jet, inlet tube, and the optical configuration shown in yellow. Sensors are shown in magenta, the water jet tube in blue ($\rightarrow$), and the flow chamber in green. The right-handed axes convention used to define the coordinate configuration of the electromagnetic coils that control our background field is also shown.

Data Collection

With the high density of NV^- centers in this diamond sample and ample laser power, the photodiode's collection is quickly saturated and acquisitions can generally be performed quite quickly ($O(\text{ms})$ per single sweep). However, given the mechanical vibrations of the pump, jet, and technical noise of the system, the total noise floor remained quite high, requiring many spectra to be averaged to produce a final resonance spectrum (300–400 spectra per 10–12 s). While this limits relevance to AC field detection, our focus is the DC/magnetostatic regime, so this compilation approach is suitable for our purposes. The oscilloscope was configured to record 10 s

records of data comprising a series of 40 ms frequency sweeps, which are compiled to a complete final spectrum and mapped to the input frequency on the basis of time. The process is repeated for each condition of the input pressure (velocity range $\approx 115\text{--}150\text{ m s}^{-1}$) and jet position. This super-cycle is repeated at various intervals to acquire the final series of spectra. Only the serial spectra within a single record are averaged prior to the fitting process.

Data Fitting and B Calculation

Resonance peaks, which appear as suppression of the NV^- center fluorescence, assume a Lorentzian-like profile. To extract eigenfrequencies, we define eight local fitting regions, one for each peak, such that they are non-intersecting. A low-order spline is first fit to the raw data, which allows estimation of the photoluminescence minima, which are taken to be the centers of each fitting region. Then, the roots of the second derivative of the spline are extracted to establish inflection points adjacent to these estimated minima. For each peak, the relevant fitting region is defined between these adjacent inflection points. The raw data, low-order spline, spline second derivative, and resulting fitting regions are shown in Fig. 4.9.

A basin-hopping algorithm is established with the known background field $\vec{B}_0$, based on Eq. 4.3, to generate synthetic spectra. The eight resonance frequencies that provide the best fit are then optimized by this algorithm via Eq. 4.3. With the known magnetic field offset used as the initial guess ($\vec{B}_0$), the fitting algorithm efficiently reaches a global solution.

Helmholtz Coil Calibration

A right-handed three-axis Helmholtz coil configuration provides a means to manipulate a bias field to separate the ESR peaks as well as a means of calibration against known B fields. With a center current of $\{-1.14, 1.80, -2.69\}$ A, corresponding to the bias field $\vec{B}_0 = (-418, 782, -2283)\text{ }\mu\text{T}$ used in this study, ODMR was applied with applied currents from 0 to 80 mA for each vector direction (xyz). Fig. 4.7 illustrates the response of resonance peaks with increasing current, and thus increasing or decreasing applied field.

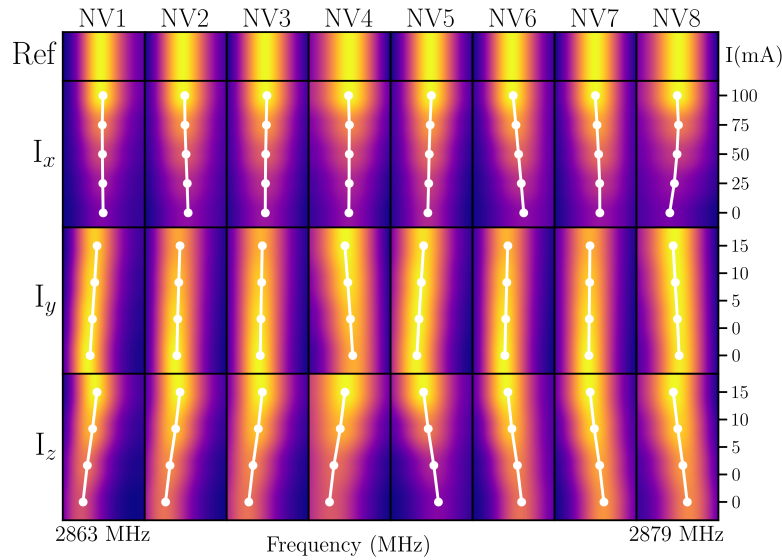


Figure 4.7: Trajectories of the eight ESR resonance peaks (columns NV1–NV8) as a function of the applied bias current along each Helmholtz coil axis (I_x , I_y , I_z , rows), with each axis scanned independently. The top row shows the reference spectrum at zero applied current. The eight peaks correspond to the four NV^- -center lattice orientations under the ± 1 spin states.

The coil parameters used for the theoretical field calculation are listed in Table 4.3. The on-axis field per unit current for a circular loop pair of radius R separated by distance L is

$$\frac{\partial B}{\partial I} = \frac{\mu_0 N R^2}{(R^2 + (L/2)^2)^{3/2}}, \quad (4.11)$$

where N is the number of turns per coil.

Table 4.3: Measured Helmholtz coil parameters, calculated on-axis field sensitivity, and measured fit slopes from coil calibration.

Orientation	N (turns)	R (mm)	T (mm)	L (mm)	$\partial B / \partial I^\ddagger$ ($\mu\text{T A}^{-1}$)	Fit slope ($\mu\text{T A}^{-1}$)	Error (%)
$\hat{x}$	40	76	6	97	396	366.59	−7.43
$\hat{y}$	40	60	6	67	557	434.90	−21.92
$\hat{z}$	40	61	6	25	800	848.91	+6.11

[‡] Parameter computed via Eq. 4.11.

From the measured values of these hand-wound and mounted coils, we estimate theoretical parameters of 396, 557, and 800 $\mu\text{T A}^{-1}$ for the x , y , and z coils, respectively.

Coil Alignment Matrix and Orthogonality Analysis

For our tests aligned along each of the x , y , and z axes, the effective coil coefficients are listed in Table 4.3 as the measured Helmholtz-coil parameters for the on-axis fields, giving respective errors of -7.43% , -21.92% , and $+6.11\%$ from theory for the x , y , and z axes, respectively. The on-axis sensitivity measurements vs. applied current are shown in Fig. 4.8.

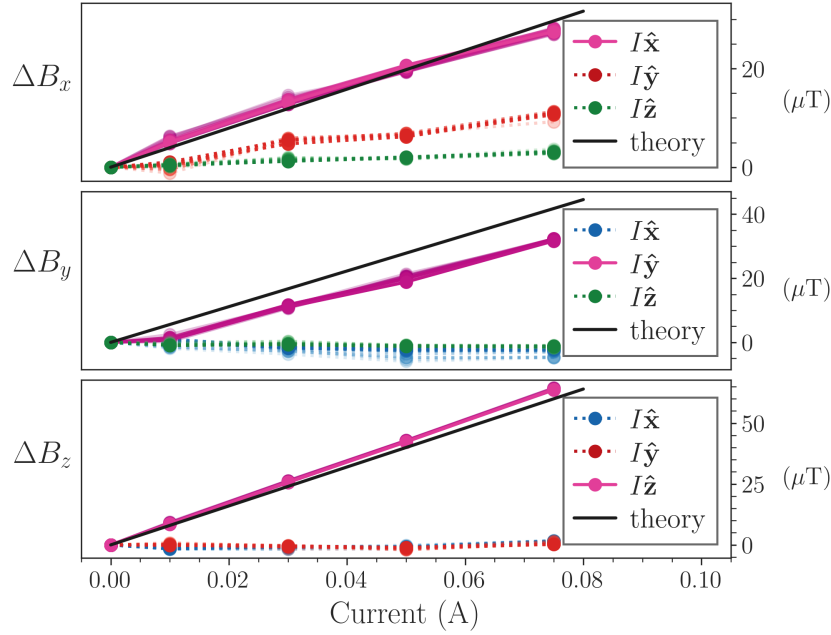


Figure 4.8: Validation measurements of the background magnetic field imposed by the three-axis electromagnetic coils, with input currents from 0 to 80 mA applied on each axis independently. The field components are shown as a function of the current applied, independently, along each respective axis. The theoretical field strength calculated from approximate coil dimensions is shown for each axis.

Since we can recover full vector projections, the full transformation from orthogonally applied currents to the measured magnetic field can be represented by the 3×3 transfer matrix $\underline{M}$:

$$\vec{\mathbf{B}} = \underline{M} \vec{\mathbf{I}}, \quad \underline{M} = \begin{pmatrix} 366.59 & 144.75 & 39.58 \\ -32.16 & 434.90 & -13.34 \\ 20.44 & -0.46 & 848.91 \end{pmatrix} \mu\text{T A}^{-1}, \quad (4.12)$$

Here $\vec{\mathbf{I}} = (I_x, I_y, I_z)$ is the applied-current vector, whose Cartesian components are the *total* currents driven along each coil axis; equivalently, for a current of magnitude I directed along polar and azimuthal angles (θ, φ) , $\vec{\mathbf{I}} = I (\sin \theta \cos \varphi, \sin \theta \sin \varphi, \cos \theta)$.

Thus $\vec{\mathbf{I}}$ is a directed total current, distinct from a current density $\vec{\mathbf{J}}$. The diagonal elements of $\underline{\mathbf{M}}$ correspond to the on-axis response for each coil, and the off-diagonal elements reflect off-axis projection from the expected xyz axes.

Given that the diamond sample is a single-crystal lab-grown diamond, the most likely source of the off-diagonal elements in $\underline{\mathbf{M}}$ is errors in coil winding or alignment, or induced magnetic field gradients from highly permeable nearby structures such as the steel optical table or steel components as part of the setup. While this measurement does not allow us to perfectly decouple intrinsic errors in the diamond crystallography, it does show that our field alignment is close to expected within a few percent. Table 4.4 summarises the tilt of each output vector from its theoretical basis, and the pairwise angles between measured axes and the resulting deviation from orthogonality.

Table 4.4: Orthogonality analysis of the measured coil basis. Top: tilt of each measured axis from its ideal lab-frame counterpart. Bottom: pairwise angles between measured axes, with deviation from 90° in parentheses.

Comparison	Angle (deg)
<i>Tilt from ideal axis</i>	
$\hat{\mathbf{e}}_x^{\text{meas}}$ vs. $\hat{\mathbf{x}}$	5.93
$\hat{\mathbf{e}}_y^{\text{meas}}$ vs. $\hat{\mathbf{y}}$	18.41
$\hat{\mathbf{e}}_z^{\text{meas}}$ vs. $\hat{\mathbf{z}}$	2.82
<i>Pairwise angles between measured axes (deviation from 90°)</i>	
(x, y)	76.63 (−13.37)
(x, z)	84.08 (−5.92)
(y, z)	90.07 (+0.07)

4.4 System Characterization

Resonance Spectrum

A representative resonance spectrum spanning ~ 2.79 – 2.94 GHz is shown in Fig. 4.9. This spectrum reveals a characteristic peak width (full-width at half-maximum; FWHM) of $\Delta f \approx 1.8$ MHz, and maximum contrast of $\approx 1.6\%$. For a single NV^- center, a maximum contrast is given by J. M. Barry et al. (2016) to be 30%. For measurements in which all four NV^- center alignments contribute to the measured PL, such as in this work, this value is proportionally averaged to 7.5%. We therefore achieve about 21% of the theoretical contrast maximum without further optimization.

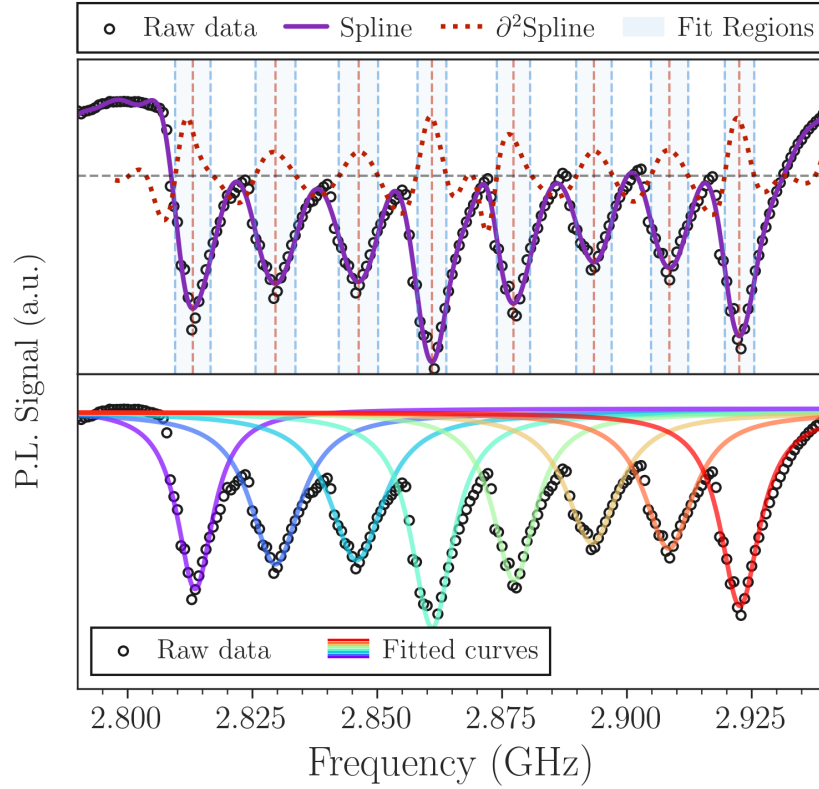


Figure 4.9: Photoluminescence spectra and fitting elements. (Top) Raw data, low-order fit spline, and the employed fitting regions. (Bottom) Eight independent fitted curves (rainbow) for the peaks comprising the final spectrum, shown overlaid on the raw data.

Sensitivity and System Noise

The sensitivity limit due to photon shot-noise in CW mode is given by J. M. Barry et al. (2016) and J. F. Barry et al. (2020)

$$\eta_{cw} = \frac{4}{3\sqrt{3}} \frac{1}{\gamma_e} \frac{\Delta\nu}{C_{cw}\sqrt{R}} \approx 9 \text{ nT Hz}^{-1/2} \quad (4.13)$$

which assumes a Lorentzian profile. γ_e is the electron gyromagnetic ratio, $\Delta\nu$ is the resonance linewidth, C_{cw} is the PL contrast factor, and R is the photon count rate. Experimentally, sensitivity is reduced by system technical noise (laser and MW power, mechanical vibrations) and environmental noise (temperature, RF contamination, etc.). Further technical noise in this setup includes primarily transient mechanical vibration, laser high-frequency noise, and drift.

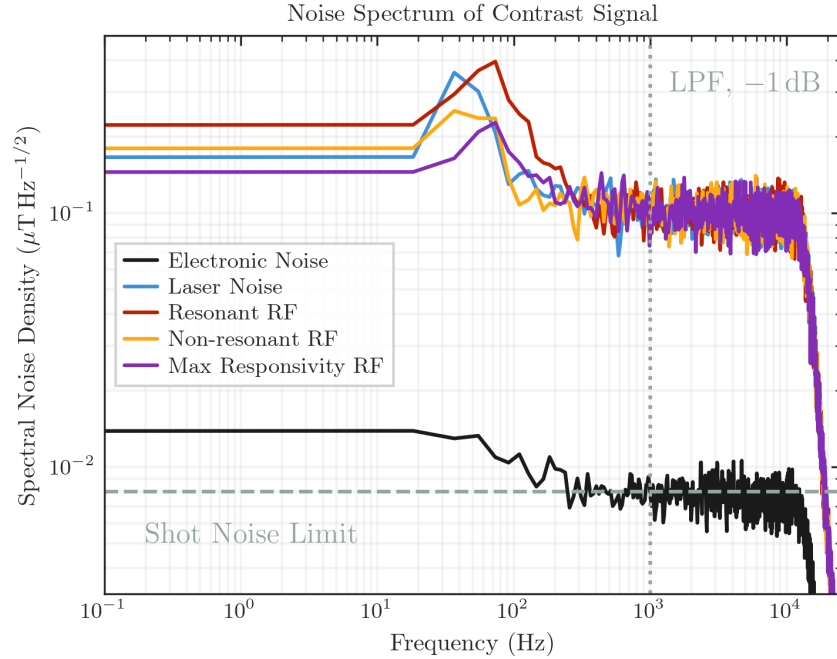


Figure 4.10: Power spectral density of system elements: max responsivity, non-resonant, and resonant spectra represent measurements integrating full RF and laser power under test conditions. Laser noise is taken without RF and electronic noise omits both laser and RF input power. The theoretical noise floor (shot-noise limited) and the low-pass filter -1 dB cutoff limits are shown for reference.

In this CW configuration, the sensitivity limits are dominated by resonant technical noise, electronic noise, and photon shot noise, in order of decreasing impact, resulting in an ultimate sensitivity of $120 \text{ nT Hz}^{-1/2}$. Fig. 4.10 shows the power spectral density in equivalent units for magnetic and electric field sensitivity.

4.5 Experimental Results

Flow Measurements

Magnetic field measurements in the near-field of the flow and plasma system were carried out along a measurement plane positioned at $\Delta z = 75 \text{ } \mu\text{m}$ below the liquid-quartz interface. A piezo translation stage traversed a span in the $\hat{x}$ direction over a domain of $x \in [-350, +350] \text{ } \mu\text{m}$. The measurement domain, measurement plane, and interrogation region size are illustrated to scale in Fig. 4.11. Relevant dimensions are shown with respect to an illustrated plasma cross-section (a), and in comparison to an image of the projected plasma geometry (b).

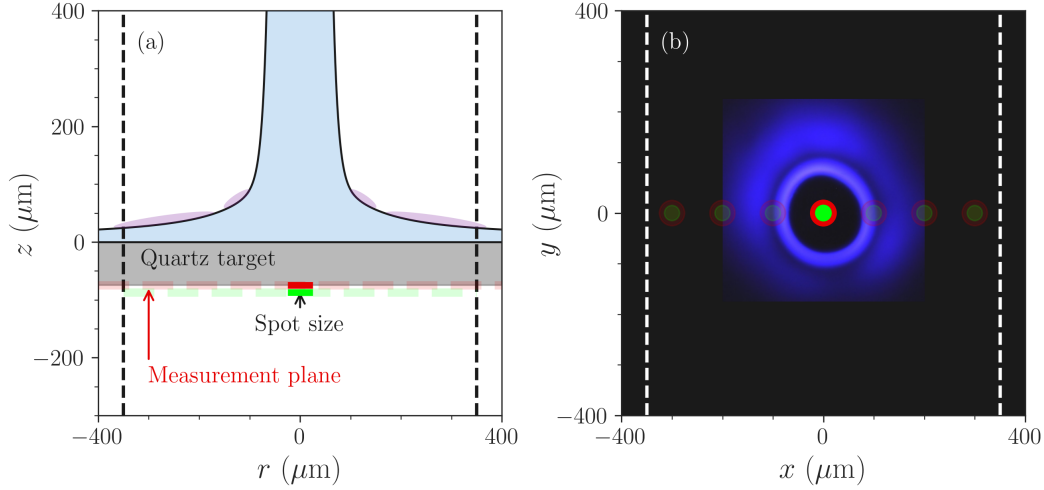


Figure 4.11: Measurement domain illustration. (a) Transverse cross-sectional view (z - x plane) showing the jet (blue), plasma (purple), the measurement plane (red and green dashed), and the interrogation spot size (●). (b) Axial view (y - x plane) of the air plasma on quartz at the same scale, showing the plasma toroidal cross-section.

The resulting measured magnetic field profiles of B_y , perpendicular to the translation direction $\hat{x}$, are shown in Fig. 4.12 for various jet velocities, as tested in tap water (top), DI water at low pressure (middle), and DI water at high pressure (bottom). Positions of measurement with respect to the jet core and plasma annulus are illustrated via pale blue circles (jet) and purple ring (plasma) in the respective panels. For all measurements, the absolute variance is $\sigma_{B_y} \approx 0.5 \mu\text{T}$, but the standard error of the mean, $\sigma_{B_y}/\sqrt{N}$, is $\sigma_{sem} \approx 10 \text{ nT}$ due to the number of records taken for each point ($N = 2000$).

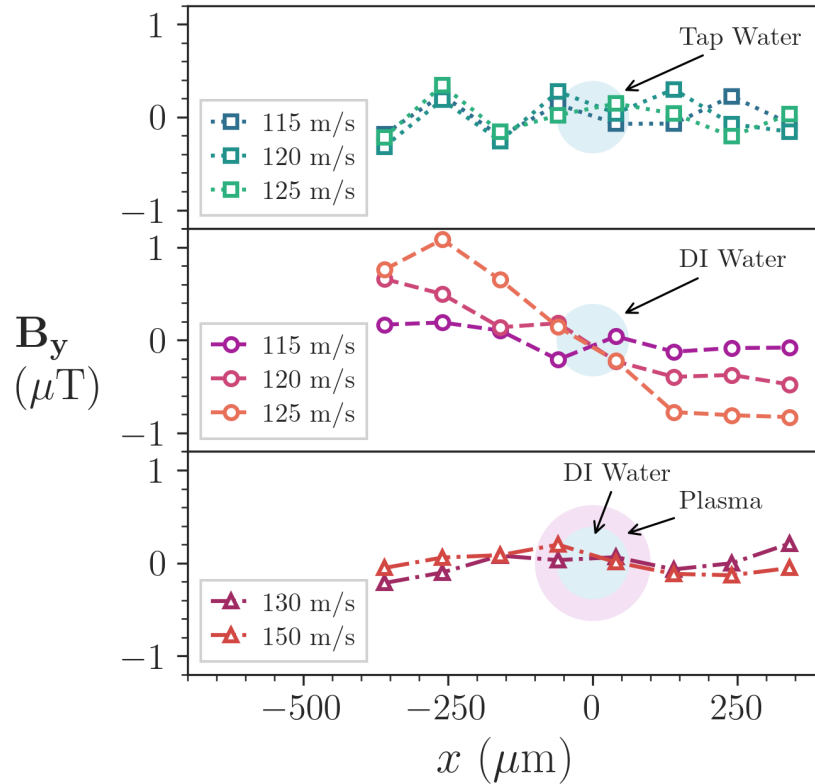


Figure 4.12: B_y as measured for a series of indicated velocities, for tap water (top), DI water at moderate pressure (middle), and DI water at elevated pressure with plasma onset (bottom). For the tap and middle DI water cases, the plasma is not seen, either a consequence of low conductivity (tap water) or low pressure (DI water). The jet core, relative to these measurements, is illustrated in blue ($\bullet$), and the presence of plasma is indicated with the corresponding purple ring ($\circ$).

From these data, we see that the tap water measurements, corresponding to the absence of plasma, show a velocity-independent B_y measurement of $\leq \pm 0.5 \mu\text{T}$, whereas in the case of distilled water we see a velocity-dependent coupling generating an antisymmetric B_y measurement of amplitude up to $\pm 1 \mu\text{T}$ at 125 m s^{-1} . Before plasma onset, the measured B_y demonstrates a velocity-dependent amplitude with linear fit of $0.075 \mu\text{T} (\text{m s}^{-1})^{-1}$, with zero-crossing at 115 m s^{-1} (control/background condition). However, as the velocity is increased past this threshold and plasma onset is achieved (around 130 m s^{-1}), the observed coupling to the azimuthal magnetic field ($B_y \approx B_\theta$) is substantially decreased. This decrease, to about $0.25 \mu\text{T}$ persists for the tested conditions of 130 and 150 m s^{-1} , which contrasts starkly against an extrapolated hypothetical value of $B_\theta(r = \pm 350 \mu\text{m}, v = 150 \text{ m s}^{-1}) \approx 2.6 \mu\text{T}$. Despite this decrease, it still manifests a structure similar to the DI-pre-plasma con-

dition. The relative position of the measured values and the jet core is shown in Fig. 4.13, and for the elevated-velocity DI water measurements, the position of the plasma core is also illustrated (shaded in purple). For clarity, these windows are omitted.

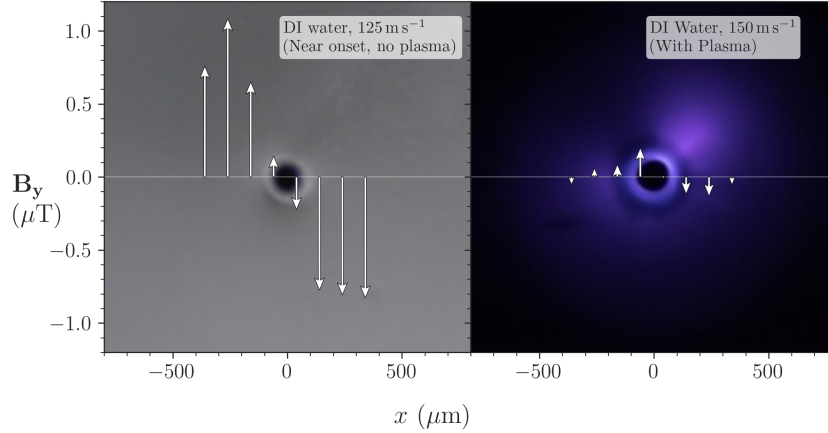


Figure 4.13: B_y measurements for the sub-plasma and plasma cases, overlaid over the jet geometry. (Left) DI water at 125 m s^{-1} (no plasma; near plasma onset); the jet core is imaged with external lighting. (Right) DI water at 150 m s^{-1} with plasma onset on quartz.

The weak coupling of tap water to the B_y field is congruent with our supposition that the conductivity of contaminated water allows faster diffusion of charge than can be sustained by any triboelectrification or streaming potential. For typical values of tap water used in this experiment, a resistivity of about $0.1 \text{ M}\Omega \text{ cm}$ was measured. In contrast, the DI water used in this experiment was typically at least $8 \text{ M}\Omega \text{ cm}$ resistivity or greater, with a theoretical resistivity of pure DI water of $18 \text{ M}\Omega \text{ cm}$.

For the signal observed with DI water, the orientation of the B_y field vectors is consistent with a flow of electrons or negative charges coupled to the flow, resulting in $B_y < 0$ at positive x and $B_y > 0$ at negative x . The observation that this occurs with DI water in a velocity-dependent manner confirms that wall-shear stress is a prerequisite for the charge generation of our system. However, the decrease in the measured values of B_y after plasma onset is seemingly contradictory to this. To explain why we do not measure further coupling to B_y after plasma onset, we conjecture this to be a natural result of pulsatile discharge behavior in the plasma which is supported by wide $\sim \text{kHz}$ frequency content of optical emission. If we describe this system in two phases, there should necessarily be a phase of charge buildup via triboelectrification, and a phase of charge depletion once

the breakdown threshold is reached. The charge buildup phase parallels our pre-discharge measurements. The discharge phase, which can be $O(\text{ns})$ at atmospheric pressure, must then have an associated current higher than the charge buildup phase, since charging is rate limited by the jet. The proportion of discharge current to charge buildup current is roughly equivalent to the inverse proportion of their timescales; the pulsed luminescence of Fig. 5.3 (Chapter 5) makes this duty-cycle relationship explicit. The observed coupling is then likely a nonlinear average of this dynamic process.

There are two main possibilities for this nonlinearity. The first is given by Eq. 4.3, which intrinsically has nonlinear terms for sufficiently high coupling. If the current is high enough to push us out of the linear regime, we expect a nonlinear average; however, the coupling retains its sign, so we would still expect an increase in the projection to B_y based on this alone.

The second possibility is that the high currents of the discharge produce instantaneous fields that push the resonance frequencies of our NV^- centers beyond the dynamic range of our system. Since we scan the resonance window from ~ 2.79 to 2.94 GHz ($\Delta f_{\text{scan}} \approx 150 \text{ MHz}$), an instantaneous magnetic field of $\Delta f_{\text{scan}}/\gamma \approx (150 \text{ MHz})/(28 \text{ MHz mT}^{-1}) \approx 5.4 \text{ mT}$ would split all resonance peaks beyond our detection region. Since our approach here has focused on magnetostatic measurements, this would intrinsically lead to broadened resonance peaks shifted to an underestimated degree. However, since we are blind to the AC component of these fields, this explanation remains primarily conjecture.

4.6 Conclusion

We have demonstrated a method for assembling an NV^- -center-based layered magnetometer which can be integrated into the walls of flow chambers and withstand megapascal-order shear stresses, placing the sensing units (the NV^- -center layer) within about $75 \mu\text{m}$ of the flow. To the best of our knowledge, this is the first such application for flow measurement based on emergent bulk magnetism. This proximity could likely be decreased further with thinner copper foil (or omission of the foil entirely), and with more precise engineering of the layer alignment and adhesive used. Proximity below this could prove useful for detection of microfluidic systems with feature sizes at or below $75 \mu\text{m}$. Further, since our microwave antenna resonance region is circular, this approach could readily be adapted to wide-field magnetic imaging with appropriate selection of cameras. Other papers (Sengottuvel

et al., 2022; Basso et al., 2025; Scholten et al., 2021) have demonstrated that wide-field magnetic imaging is possible and suggested specific hardware configurations that might achieve this; however, to the best of our knowledge, this has not been applied to imaging of flow structures through magnetic coupling.

In addition to expanding the spatial domain, there is great potential for accessing the temporal domain for both low- and high-frequency electromagnetic sensing. This could be accomplished through a combination of dynamic microwave excitation pulse sequences and extension of this technique to Hahn-echo decay or Rabi pulse-sequence sensing as commonly used in MRI, for substantial increases in fidelity across spatio-temporal measurement domains.

4.7 Summary

Micron-scale toroidal plasmas generated purely through water-solid triboelectrification exhibit extreme shape stability, temporal stability, and symmetry that are atypical of laboratory plasmas. The cylindrical symmetry and radial segmentation of this plasma type strongly suggest intrinsic quasi-steady behavior (see the Gallery, images AQ8, AL3, AL7, AL9, ArQ2, ArQ4, ArL1–ArL3, HeQ1, and HeL1). Previously (see Chapter 1), the electric field near this plasma was mapped, demonstrating a poloidal structure consistent with a negative coronal discharge and a steady-state poloidal current. Additionally, in the case of an argon plasma (see Chapter 2), the visible luminescence has been spectroscopically probed to determine that the inner stable core region, which persists in all gases, exists with an elevated temperature and depressed electron density, with a stark boundary from the diffusive plasma exterior. We postulate that this current is accompanied by a self-generated azimuthal magnetic field whose role in the plasma’s coherent toroidal structure—whether reinforcing, negligible, or even destabilizing—remains to be determined (see Chapter 5). Due to its small scale and symmetry, far-field measurements ($r > 100 \mu\text{m}$) are inadequate. To probe the magnetic signature of these plasmas, this work designs a wall-integrated magnetometer based on nitrogen-vacancy centers in diamond. These atomic-scale fluorescent defects have energy levels that are highly sensitive to local magnetic fields. By layering an ultra-thin ($50 \mu\text{m}$) quartz target wafer, thin copper foil ($10 \mu\text{m}$), and a surface-NV⁻-doped ($5 \mu\text{m}$) diamond within the wall of the plasma’s generating flow, we demonstrate a near-field (sub-length-scale, here $\sim 75 \mu\text{m}$) sensor format that can be non-invasively applied to micron scale flows by integration into the flow wall. Due to the fundamental dipole nature of magnetic field structures, proximity is an essential component of small-scale B -field

measurements. Under continuous-wave excitation, we achieve with this method a shot-noise-limited sensitivity of $9 \text{ nT Hz}^{-1/2}$ and a technical-noise-limited sensitivity of $120 \text{ nT Hz}^{-1/2}$. In the case of extreme wall shear in the near-field of an impinging water jet, we observe the signature of an antisymmetric current element (measured at $z \approx -75 \text{ }\mu\text{m}$) below plasma onset with an amplitude of $\approx 1 \text{ }\mu\text{T}$. After plasma onset, this value drops to $\approx 0.25 \text{ }\mu\text{T}$. The polarity of this structure is congruent with a surplus of electrons being advected by the flow. We further posit that the onset of plasma induces an unsteadiness which, due to the intense averaging used by this particular sensing configuration, pushes measurements outside the dynamic range of the sensor.

Chapter 5

MODELLING THE ELECTRIC AND MAGNETIC FIELD

We have demonstrated a method that confirms the existence of an asymmetric magnetic field measured within $75\text{ }\mu\text{m}$ of the jet–flow-wall interface under the condition of high-speed flow of pure water, only in the absence of plasma. The predicted plasma-associated field was not observed. This asymmetric structure of the pre-discharge state (flow without plasma) is congruent with the presence of a poloidal current advected with the jet, generating an azimuthal B_θ ($|B_\theta| \equiv |B_y|$ at $x = 0$) coaxial with the flow — present only within the region that encloses the current. As the jet flow is the underlying source of this equivalent electric current, we assume that this source is, like the jet, axisymmetric. While various non-symmetric plasma modes exist (see the Gallery, images AQ3–AQ5, AL9, etc.), the lowest order mode appears nearly perfectly symmetric (see the Gallery, images AQ6, AQ8, AQ9, ArQ1, ArQ4, HeQ1, and HeQ4).

Crucially, our measurement is made *outside* the enclosed current (below the target plate): for a perfectly axisymmetric closed current, Ampère’s law gives *no* azimuthal field in this external region. The finite measured field $B_y(\pm 350\text{ }\mu\text{m}) \approx \pm 1\text{ }\mu\text{T}$ therefore reflects the *non-axisymmetric* component of the current distribution—we are sensing departures from perfect axisymmetry rather than the idealized enclosed-current field. To estimate this internal B_θ (which corresponds to the inner energetic core of this plasma described in Chapter 2), we develop a simple electrostatic and magnetostatic picture on the basis of plasma topology, plasma parameters, and the triboelectrification model described in Chapter 1. First, by defining a negatively charged region (cathode) and a positively charged region (anode) and solving the axisymmetric form of Gauss’s law, we compute a proposed distribution of the electric potential $\phi(r, z)$ and field $E(r, z)$. Then we treat the magnetic field of the associated electric current using Ampère’s law: a closed, axisymmetric (poloidal) current encloses a net current I , and produces an azimuthal field

$$B_\theta(r, z) = \frac{\mu_0 I(r, z)}{2\pi r}, \quad (5.1)$$

where $I(r, z)$ is the current enclosed by a circle of radius r at height z . The field vanishes wherever no net current is enclosed (for example, below the substrate, or

beyond a closed circuit whose return path is also enclosed).

We consider two currents within this single closed circuit. The first is a weak *fluid charging current* $I_F \sim O(10 \text{ mA})$, advected with the jet in the pre-discharge phase; this is the current responsible for the field we measure without plasma. The second is the much larger, transient *discharge current*, with an instantaneous peak $I_D \sim O(50 \text{ A})$ present only during the active discharge. Both magnitudes are highly uncertain order estimates, and we carry their stated assumptions explicitly throughout. The charge advected by the jet need not imply that the bulk fluid carries a large net charge: the triboelectric mechanism of Chapter 1 requires only that charge be separated at the shear interface (the fluid may become progressively less negative as it deposits electrons on the surface), and the magnetic picture below holds regardless of the sign convention. As we show below, the azimuthal field of such an enclosed current exerts an outward magnetic pressure ($B_\theta^2/2\mu_0$) that tends to *expand* the current loop, with a magnitude comparable to the plasma electron pressure.

The following analyses assume an axisymmetric system with continuous rotational symmetry ($\vec{\mathbf{E}}(r, \theta, z) = \vec{\mathbf{E}}(r, z)$, $\vec{\mathbf{B}}(r, \theta, z) = B_\theta(r, z) \hat{\theta}$, $\vec{\mathbf{J}}(r, \theta, z) = \vec{\mathbf{J}}(r, z)$) with central axis $\hat{\mathbf{z}}$, unless explicitly noted otherwise. Non-dimensional spatial variables

$$\mathcal{R} \equiv \frac{r}{R}, \quad \mathcal{Z} \equiv \frac{z}{R} \quad (5.2)$$

are defined with respect to the jet radius $R \equiv 62.5 \mu\text{m}$, at impingement velocity $v = 125 \text{ m s}^{-1}$ and 40°C , with $Re \equiv Ul/\nu \approx 30,000$ and $We \equiv \rho U^2 l / \sigma \approx 40,000$. This velocity is about the threshold for plasma onset and is therefore a reasonable baseline within measurement error.

5.1 Electric Field Model

Chapter 1 established that electrical potential is generated via triboelectrification at the jet–target interface, proportional (to first order) to the wall shear stress τ_w (Vihacencu et al., 2014). On the basis of the location of $\max(\tau_w)$ at the interface and the plasma characteristic size ($\sim 500 \mu\text{m}$), we define a charged (pre-discharge) configuration: a negative ring near the high-shear annulus (the cathode) and an equal, opposite positive ring downstream at the liquid surface (the anode), scaled to maintain overall space-charge neutrality. The dielectric constants of the quartz substrate, water jet, and air ($\epsilon_{\text{quartz}} = 4$, $\epsilon_{\text{water}} = 80$, $\epsilon_{\text{air}} = 1$ (Uematsu and Franck, 1980; Bottom, 1972)) are imposed on the corresponding regions.

From $\rho(\vec{r})$ and $\varepsilon(\vec{r})$, the axisymmetric equation for the potential is solved in the FEniCS Project PDE solver in axisymmetric coordinates $\vec{r} = (r, z)$:

$$\frac{1}{r} \frac{\partial}{\partial r} \left(\varepsilon(\vec{r}) \frac{\partial \phi(\vec{r})}{\partial r} \right) + \frac{\partial}{\partial z} \left(\varepsilon(\vec{r}) \frac{\partial \phi(\vec{r})}{\partial z} \right) = -\rho(\vec{r}) \quad (5.3)$$

and the electric field follows as

$$\vec{E}(\vec{r}) = -\nabla \phi(\vec{r}). \quad (5.4)$$

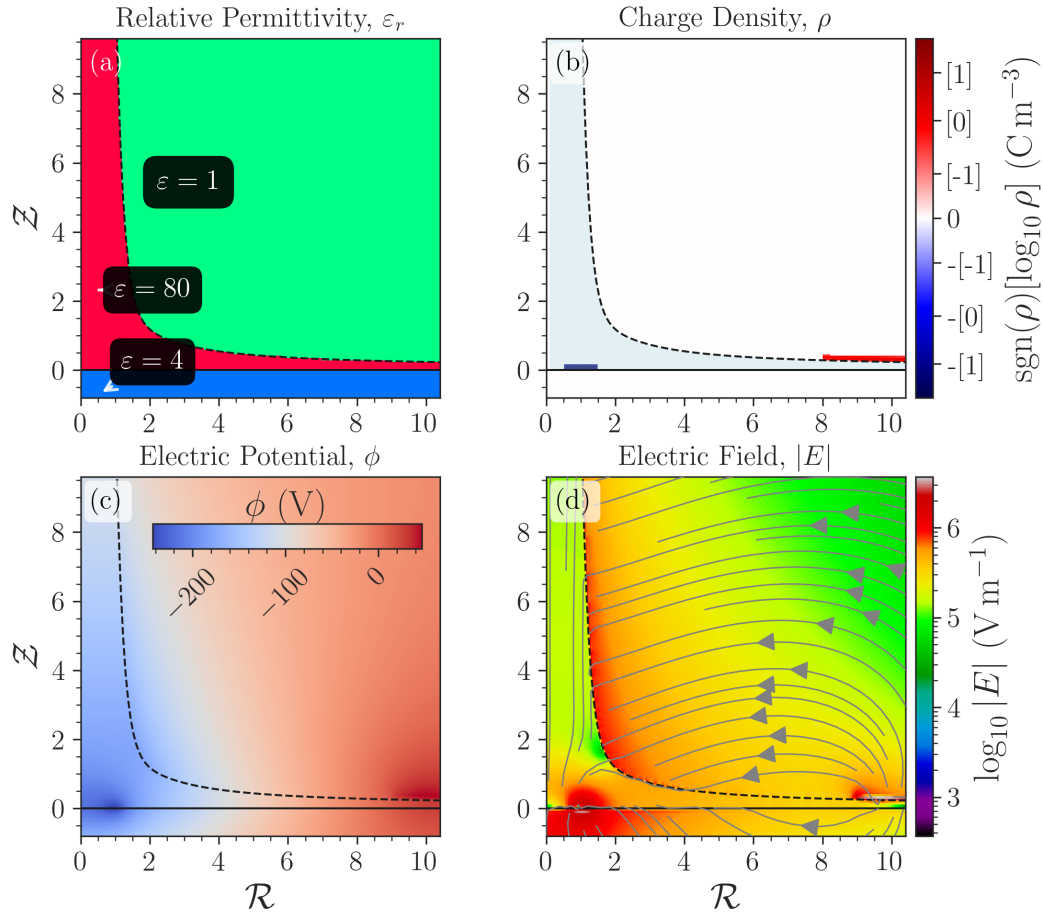


Figure 5.1: Electric field model. Panel (a): relative permittivity distribution. Panel (b): charge density distribution, with negative and positive ring charges represented by blue (■) and red (■) regions, respectively. Panel (c): resulting electrical potential $\phi(r, z)$. Panel (d): resulting electric field $E(r, z)$, with streamlines indicated in gray ($\rightarrow$).

The resulting distributions of $\phi(r, z)$ and $E(r, z)$ are shown to approximate scale (based on the measured $\max(\Delta\phi) \approx 300 \text{ V}$) in Fig. 5.1(c) and (d). We see two intense

regions of electric field corresponding to the regions of finite charge density, with the expected discontinuity at the liquid interface and a concentration around the jet annulus (the region of maximum curvature). The air side exhibits a greatly elevated electric field while the fluid side exhibits a local minimum. This distribution serves as the basis for the discharge geometry considered below.

5.2 Magnetic Field of an Enclosed Current

To approximate the magnetic field we consider two stages of this triboluminescent process. First, the pre-discharge state, where electrical potential has built but remains below the threshold for electrical breakdown; this is the state directly related to our B_θ measurements (Chapter 4, Fig. 4.12). Second, the active discharge, under the instantaneous condition that the discharge current spans the full inter-electrode gap. Bifurcating the process into two stationary phases is an oversimplification, but omitting the charging term ($\partial\rho/\partial t$) means we are estimating an upper bound on the magnetic effect, as all charge is ascribed to current flows,

$$\mu_0\epsilon_0(\partial E/\partial t) = 0; \quad \nabla \times \vec{\mathbf{B}} = \mu_0\vec{\mathbf{J}}. \quad (5.5)$$

Both currents are legs of a *single closed circuit*, shown illustratively in Fig. 5.2: the flow mechanically separates charge and carries the fluid current I_F out along the spreading film; this charge ultimately returns through the external collection/recirculation path, and during breakdown a transient discharge current I_D recombines charge locally across the gas gap. Because the circuit is closed, Ampère's law (Eq. 5.1) fixes the azimuthal field from the enclosed current alone, and no field exists in regions enclosing no net current.

The Fluid Charging Current

In the pre-discharge state, current exists only through the advection of charge carried by the jet flow; this is the regime of our plasma-free measurement of an antisymmetric B_θ (Fig. 4.12, Chapter 4). The triboelectrification of Chapter 1 separates charge at the shear interface, so the conventional current is advected with (or against) the flow; this is congruent with our measurements and with other studies (Vácha et al., 2012; Santos et al., 2011). The accompanying charging current — the slow accumulation of separated charge — is necessarily a small fraction of I_F and is neglected, since the discharge timescale ($\sim$ ns) is far shorter than the charging timescale ($\sim$ μ s); this separation of timescales is directly visible in the pulsed luminescence of Fig. 5.3.

The magnitude of this charging current is set by charge conservation: in steady

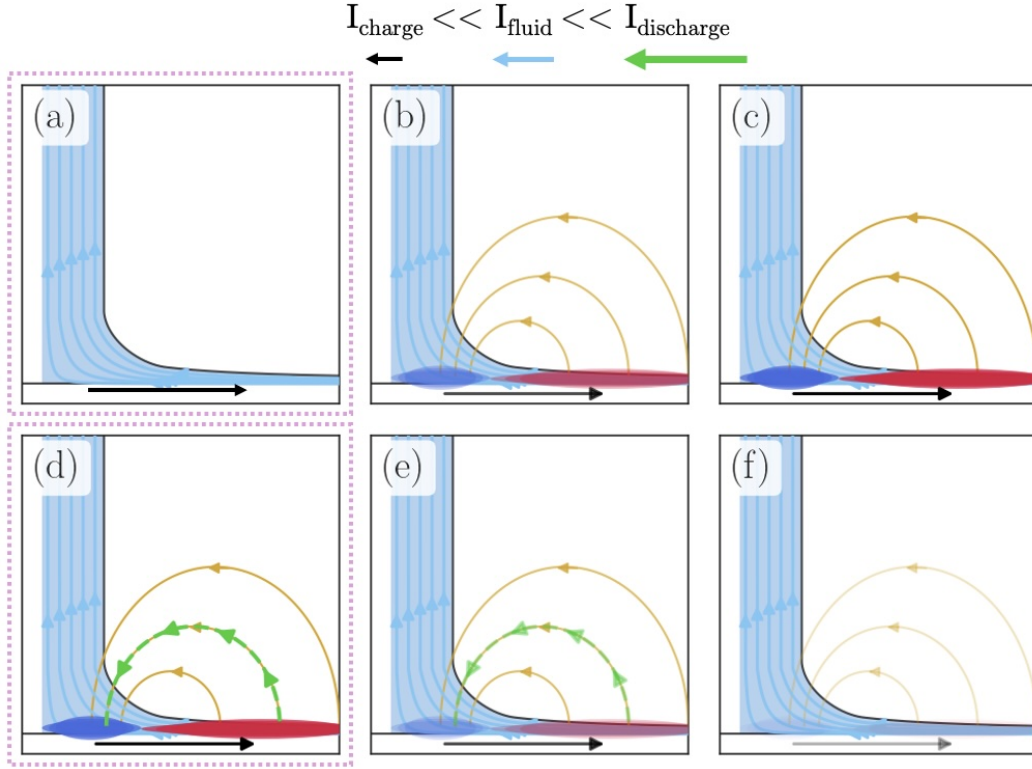


Figure 5.2: The charge/discharge cycle drawn as a single closed current circuit. The flow advects the fluid charging current I_F outward along the film (charging phase); the charge returns through the external loop; and the transient discharge current I_D closes the circuit locally across the gas gap (discharging phase). Only the passive charging and active (peak) discharging phases are considered here.

state the charge advected by the jet must equal the charge released by the discharge, so I_F is fixed by the *time-averaged* discharge current. As estimated in Section 5.2 from the pulse duty cycle (Fig. 5.3), this gives $I_F \sim O(10 \text{ mA})$ — far below the instantaneous discharge peak. For a perfectly axisymmetric enclosed current of this magnitude, Ampère’s law (Eq. 5.1) would give an azimuthal field at the core radius $r \approx 100 \text{ }\mu\text{m}$ of $B_\theta = \mu_0 I_F / (2\pi r) \approx 20 \text{ }\mu\text{T}$.

Our measurement, however, is taken below the target at a standoff $z \approx -75 \text{ }\mu\text{m}$ — *outside* the enclosed current — where a purely axisymmetric closed current produces no azimuthal field. The finite signal we record ($\pm 1 \text{ }\mu\text{T}$) therefore arises from the *non-axisymmetric* component of the current and probes departures from perfect axisymmetry, rather than the idealized enclosed-current field.

The Discharge Current

Our measurements show B_θ prominently in the charged (pre-discharge) state, and a much smaller signal after plasma onset. Given the transient nature of atmospheric-pressure discharges ($O(\text{ns} - \mu\text{s})$), the equivalent current is likely concentrated into rapid charge-depletion cycles between accumulation periods. Because the dynamic range of our sensor is $O(10 \text{ mT})$, our long averaging produces a nonlinear average that, after onset, no longer represents the instantaneous current. This pulsatile behaviour is directly observed in the plasma luminescence (Fig. 5.3), where nanosecond-scale emission bursts are separated by microsecond-scale intervals.

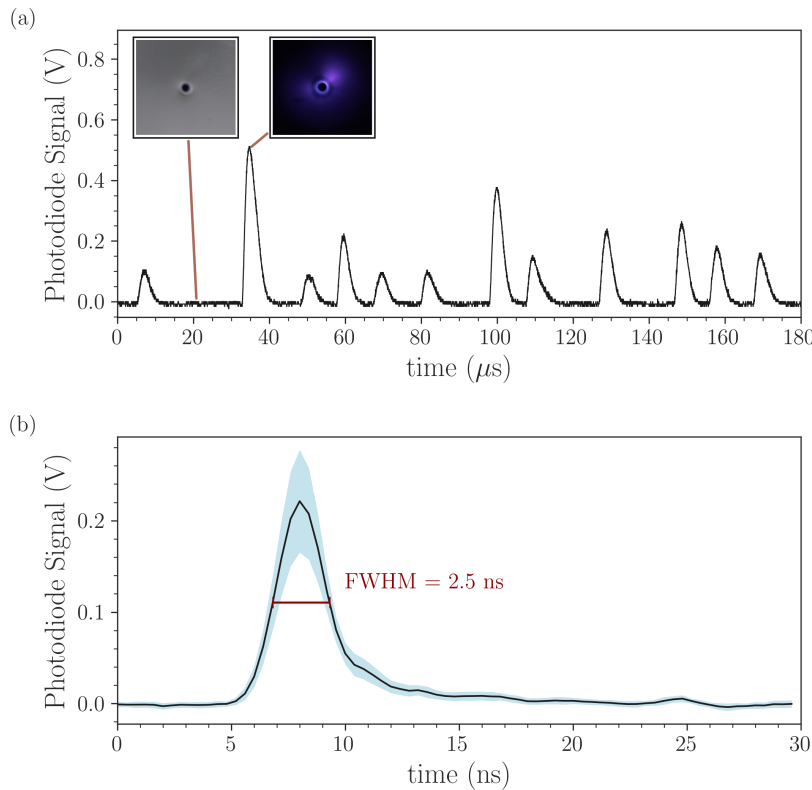


Figure 5.3: Pulsatile discharge behaviour, recorded as plasma luminescence. (a) Pulse train over $\sim 200 \mu\text{s}$, measured with a 100 kHz APD (Thorlabs APD440A2) whose limited bandwidth artificially broadens the apparent pulse widths; insets show the plasma-on (bright toroidal core) and plasma-off states. (b) A single pulse resolved with a 400 MHz APD (Thorlabs APD430A2), shown as a Gaussian-windowed average of 40 pulses, with $\text{FWHM} \approx 2.5 \text{ ns}$. The brief pulse width and $O(10 \mu\text{s})$ spacing give a duty cycle of $O(10^{-4})$, so a time-averaged (continuous-wave) measurement greatly underrepresents the peak instantaneous current and field, under the assumption that discharge current correlates exactly to luminescence intensity. This is likely a generous estimate.

For the active discharge we take a characteristic current density $J \approx 7.93 \text{ mA } \mu\text{m}^{-2}$ (calculated for the nitrogen plasma from spectroscopic data; Appendix B, Section B.1), through a plasma thickness $t \approx 10 \text{ } \mu\text{m}$ at a nominal core radius $r \approx 100 \text{ } \mu\text{m}$. The resulting *instantaneous peak* discharge current is

$$I_D = J \cdot 2\pi r \cdot t \approx (7.93 \text{ mA } \mu\text{m}^{-2})(2\pi \cdot 100 \text{ } \mu\text{m})(10 \text{ } \mu\text{m}) \approx 50 \text{ A}.$$

This peak is highly uncertain, resting on the spectroscopically inferred current density and the assumed thickness and radius. It is, however, an *instantaneous* value: Fig. 5.3 shows the discharge luminescence occurring in brief bursts (FWHM $\approx 2.5 \text{ ns}$) separated by $O(10 \text{ } \mu\text{s})$ intervals, a duty cycle of $O(10^{-4})$, assuming that discharge current correlates exactly to luminescence intensity. The *time-averaged* discharge current is therefore

$$\langle I_D \rangle \approx I_D \frac{\tau_{\text{pulse}}}{\tau_{\text{period}}} \approx (50 \text{ A}) \frac{2.5 \text{ ns}}{15 \text{ } \mu\text{s}} \approx O(10 \text{ mA}),$$

which by charge conservation matches the steady fluid charging current I_F of Section 5.2. A pulsed discharge thus reconciles a large instantaneous current with a small average: the current is momentarily high even though the time-averaged value—and the post-onset signal—is low.

By Ampère’s law the field at the current surface for the *peak* current is $B_\theta = \mu_0 I_D / (2\pi r) \approx 100 \text{ mT}$, with a magnetic pressure

$$P_B = \frac{B_\theta^2}{2\mu_0} \approx \frac{(100 \text{ mT})^2}{2\mu_0} \approx 4 \text{ kPa}.$$

The force associated with B_θ^2 is directed outward: like parallel elements of a poloidal current, the enclosed current repels itself, so the loop tends to *expand*. This expansion pressure ($\sim 4 \text{ kPa}$) is of the same order as—though likely somewhat below—the plasma electron pressure (10–68 kPa, Table 5.1); it is weaker but not negligible. Both terms push outward, so any model of the observed confinement must account for the magnetic pressure alongside the electron pressure.

5.3 Conclusions

On the basis of our measurements, the jet geometry, and an electric field model that closely parallels those measurements, we have identified an axisymmetric magnetic field connected to our flow that is congruent with a velocity-dependent charging mechanism and an associated toroidal (azimuthal) magnetic field. The measured field is present with the flow of high-speed pure water, but seemingly absent once

the velocity is sufficient to induce plasma onset; the predicted plasma-associated field was not observed. Treating the system as a single closed current circuit, the azimuthal field follows directly from the enclosed current by Ampère’s law (Eq. 5.1), with no field where no net current is enclosed.

For the fluid charging current ($I_F \sim O(10 \text{ mA})$, the time-averaged discharge current) a perfectly axisymmetric current would give a surface field of $O(20 \text{ } \mu\text{T})$; the field we actually measure outside the current ($\pm 1 \text{ } \mu\text{T}$) instead reflects its non-axisymmetric structure. For the instantaneous discharge peak ($I_D \sim O(50 \text{ A})$, highly uncertain) the surface field at $r \approx 100 \text{ } \mu\text{m}$ is $O(100 \text{ mT})$, and its B_θ^2 magnetic pressure ($\sim 4 \text{ kPa}$) acts *outward*, tending to expand the current loop. This expansion pressure is of the same order as the plasma electron pressure ($n_e k_B T_e \sim 10\text{--}68 \text{ kPa}$; Table 5.1) — likely somewhat weaker, but not negligible. Both the electron pressure and the magnetic pressure are directed outward, so neither confines the plasma on its own; a self-consistent model of the observed structure must balance both against an as-yet-unidentified inward force. As a consistency check, the hydrodynamic power flux of the jet ($\frac{1}{2}\rho U^3 \pi r^2 \approx 125 \text{ W}$ for a $100 \text{ } \mu\text{m}$ -radius jet at 200 m s^{-1}) provides an upper bound on the electrical power available to the system. The stark symmetry of the toroidal core is therefore more plausibly set by the impinging-jet stagnation hydrodynamics and the triboelectric charging chemistry than by the magnetic self-field — though that self-field is not negligible and merits inclusion in any confinement model.

The stark symmetry and unique character of the plasma core nevertheless invite the question of what role a magnetic field might play. In non-thermal, non-equilibrium atmospheric plasmas, prior experiments have shown that imposed magnetic fields in the range $20\text{--}420 \text{ mT}$ can modify plasma behaviour (Gao et al., 2019; Jiang et al., 2014; Li et al., 2022; Wang et al., 2017): most commonly increasing homogeneity and reducing filamentation (Jiang et al., 2014; Wang et al., 2017), lengthening high-aspect-ratio plasmas (Shi et al., 2023), or — in discharges with a sufficient DC current component — opposing expansion via the magnetic Lorentz term (Li et al., 2022). The $r\text{--}z$ cross section of our plasma recovers an almost identical topology to the tapered expansion discharge studied by Li et al. (2022), suggesting that magnetic influence on structure, if not confinement, remains a worthwhile question.

5.4 Limitations and Future Work

Extension of the Experiment

A critical experimental limitation borne from the need for extensive averaging is the restriction to scanning along one dimension, using a single jet size (125 μm diameter), and coarse spatial and experimental parameter discretization. These limitations offer several experimental paths forward. The first is NV^- sensing based on dynamic pulse schemes, which can offer many times higher sensitivity compared to the continuous-wave approach (Silani, 2023). Additionally, coupling this with a wide-field scan of our NV sensor, for example through the use of a lock-in camera, could provide one-shot full-frame information instead of the point-by-point approach of a scanning microscope (Wojciechowski et al., 2018). Wide-field resolution would additionally allow for the observation of the non-symmetrical plasma modes in which various streamers appear with unique geometries and odd symmetry. Furthermore, averaging has also constrained our measurements to strictly the DC regime; the pulsed and one-shot approaches offer the possibility to resolve AC signals over a wide range of frequencies. Performing the same experiments in the case of argon or helium, which have drastically different plasma parameters, would require significantly accelerating our measurement technique.

Extension of the modeling approach

Our modelling is restricted to a two-phase, magnetostatic picture: the pre-discharge (charging) regime, where the current is low and constant, and the discharge regime, with an additional superposed discharge current. This is a drastic simplification of plasma dynamics, in that we have considered the electric and magnetic fields only statically; on the basis of Maxwell's equations these fields are all but inseparable in four-space. Expanding our models to the time domain would provide more insight into the dynamic behavior and coupling between the electric and magnetic field components.

Physical analogs

A solid-state analog — a copper or otherwise conductive structure (solid or hollow, each configuration representing a different current distribution within the fluid) mimicking the jet's flow geometry — could carry a controlled current and reproduce the same measurement at a far larger scale, provided axisymmetry is broken so that the field is externally measurable; this could even be done with a standard magnetometer at the centimeter scale rather than the micron scale. Doing so would eliminate much of the experimental uncertainty. A natural variant is to increase or decrease the jet size to tune the radius of curvature while maintaining similar discharge characteristics. Another variation is a physical bulk model constructed

from quartz or ice, which would allow embedding concentric ring electrodes to create a dielectric barrier discharge on a more adaptable scale without the presence of flow. This would enable a view into the pure plasma dynamics around a dielectric structure, decoupled from the fluid flow and its associated current.

Surface interactions

Finally, one element whose role is unclear is the effective lensing through this geometry: from the model for the electric field (Fig. 5.1), the electron drift velocity V_D can be inferred (Paris et al., 2005). The 2D distribution in an r - z plane is shown in Fig. 5.4. In the liquid phase, we observe a local (almost global) minimum of V_D — coincidentally at the exact location of our inner toroid of interest. While this abstracts away the liquid aspect of this phase, this minimum is completely absent in the gas phase and is purely a consequence of the concavity of the dielectric constant of the water in this region. So far, we have assumed (and experiments have validated) that luminescence comes from the gas phase; however, the free surface interaction or the dynamics in the fluid itself may give rise to additional phenomena which have not yet been considered. Future study may consider the contribution of this node, and whether a unique surface effect between water and plasma contributes to the observed structure. A gallery of the various plasma configurations is included below for your enjoyment.

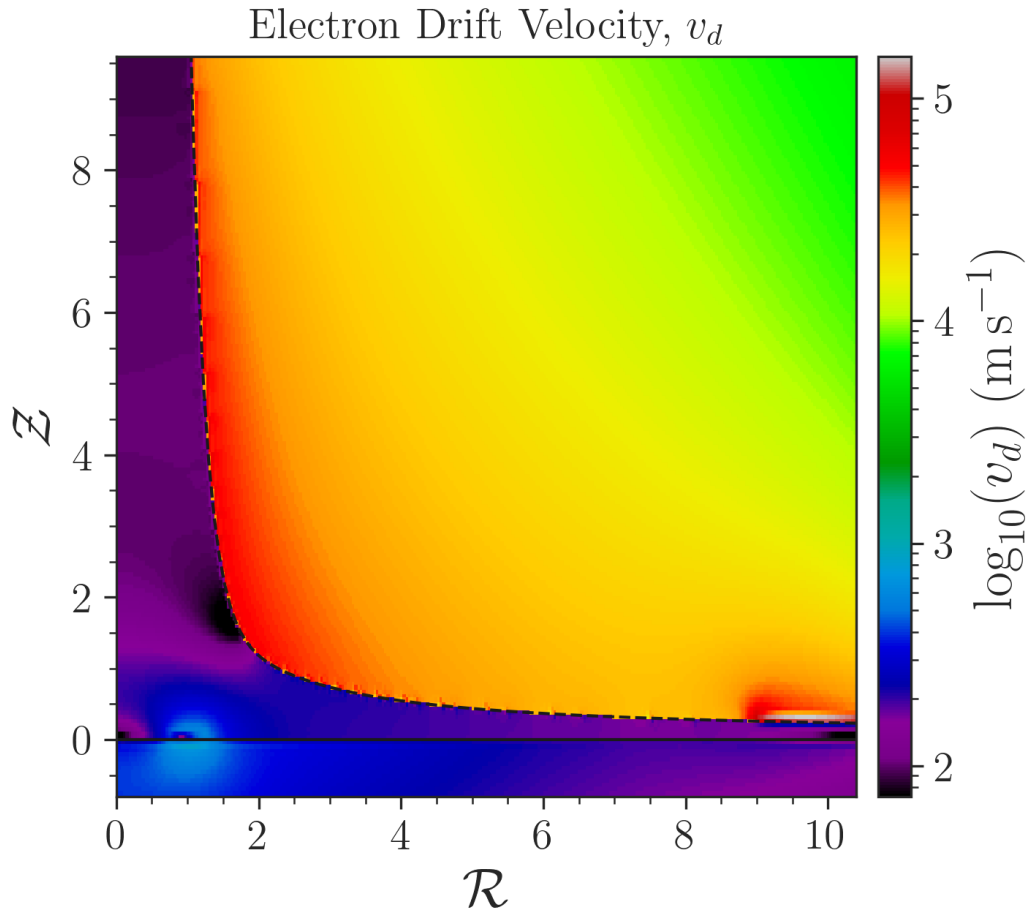


Figure 5.4: Electron drift velocity V_D , proportional to E/N , plotted on $\log_{10}$ scale against non-dimensionalized coordinates. Around $\mathcal{R} = 2$ and $\mathcal{Z} = 2$, a local minimum appears with drift velocity about 80 m s^{-1} .

Table 5.1: Plasma parameters for the three analyzed cases (N₂, He, and Ar). The bottom row gives the electron pressure $n_e k T_e$.

Parameter	N ₂	He	Ar
M_{ion} (amu)	28	4	40
n_e (m ⁻³)	10 ²² –10 ²³	— [†]	2.2×10^{21}
T_e (eV)	0.24–2.64	— [†]	1.1
T_g (K)	1200	2200	4500
f_{pe} (Hz)	1.42×10^{14}	3.50×10^{13}	3.54×10^{11}
f_{pi} (Hz)	6.27×10^{11}	4.15×10^{11}	1.31×10^9
ν_{ei} (Hz)	2.00×10^{12}	9.18×10^9	4.62×10^7
λ_D (μm)	0.03	0.3	7
δ_{skin} (mm)	4	1	7
U_i (m s ⁻¹)	6×10^2	2×10^3	1×10^3
U_e (m s ⁻¹)	7×10^5	2×10^6	4×10^5
c_s (m s ⁻¹)	4×10^3	2×10^4	2×10^3
$n_e k T_e$	10 kPa–68 kPa	— [†]	≈ 390 Pa

[†] Helium plasma parameters cannot be reliably extracted in this experimental regime: instrument FWHM dominates the available Stark-broadening line widths. See Chapter 1, Section 1.2, for details.

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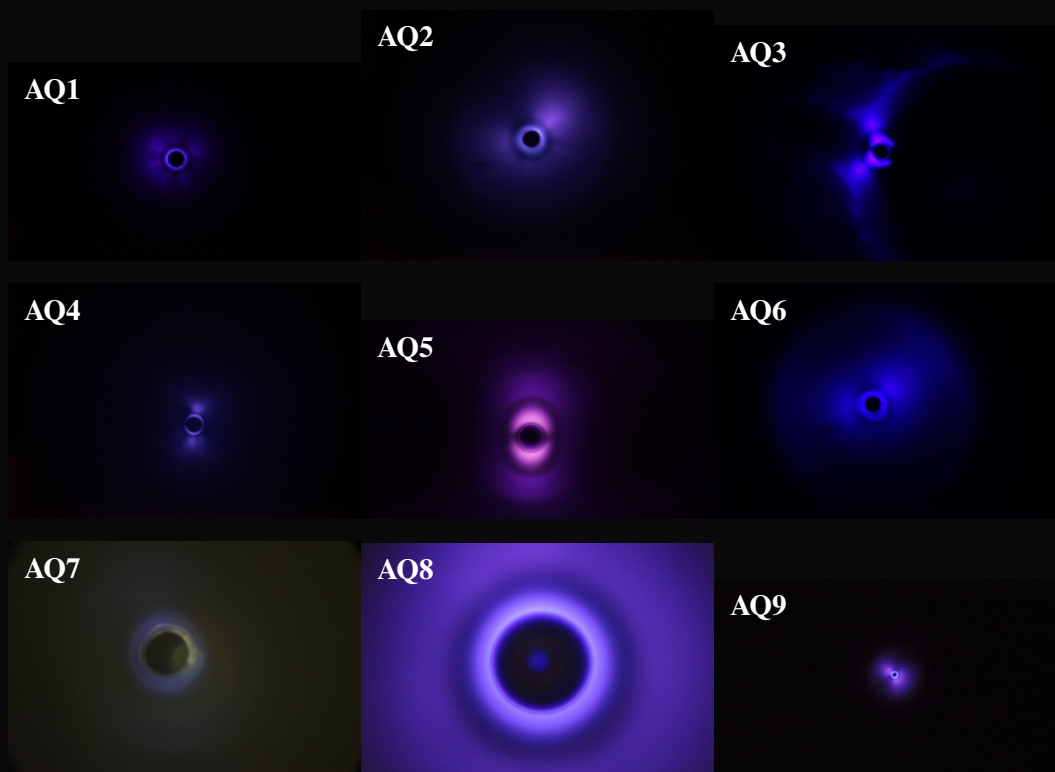
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Appendix A

PLASMA GALLERY

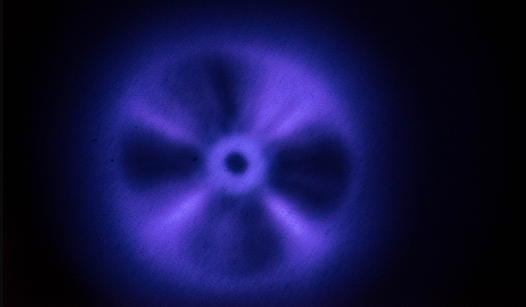
**Air
Quartz**



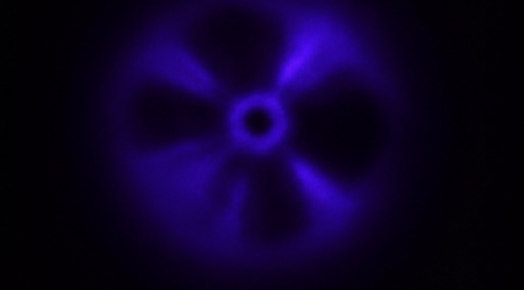
Lithium Niobate



AL3



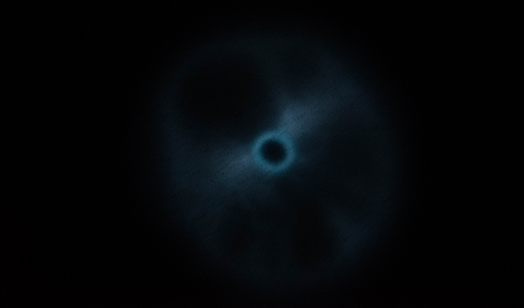
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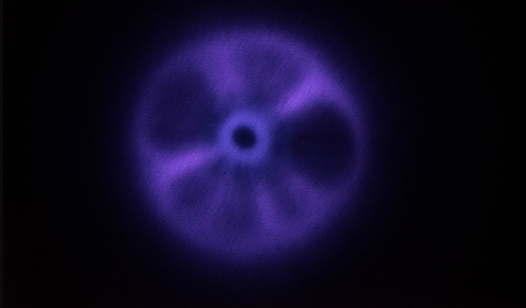
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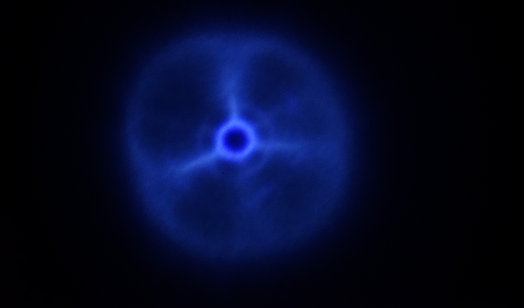
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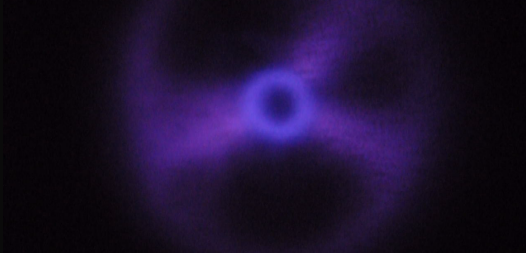
AL7



AL8



AL9

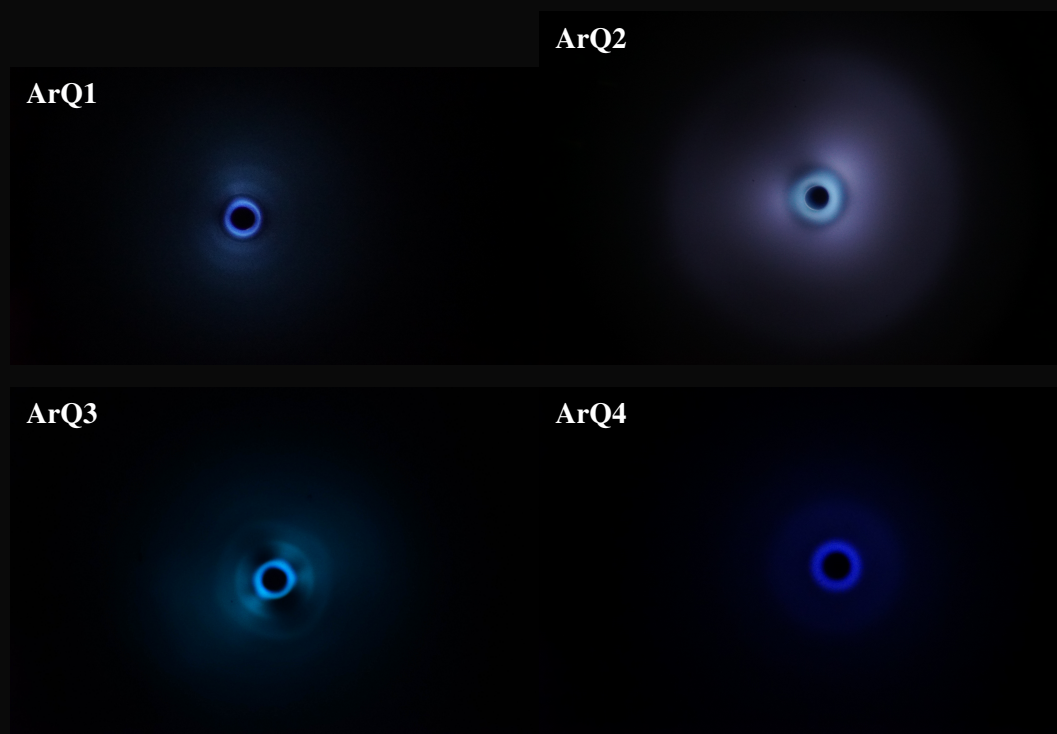


AL10

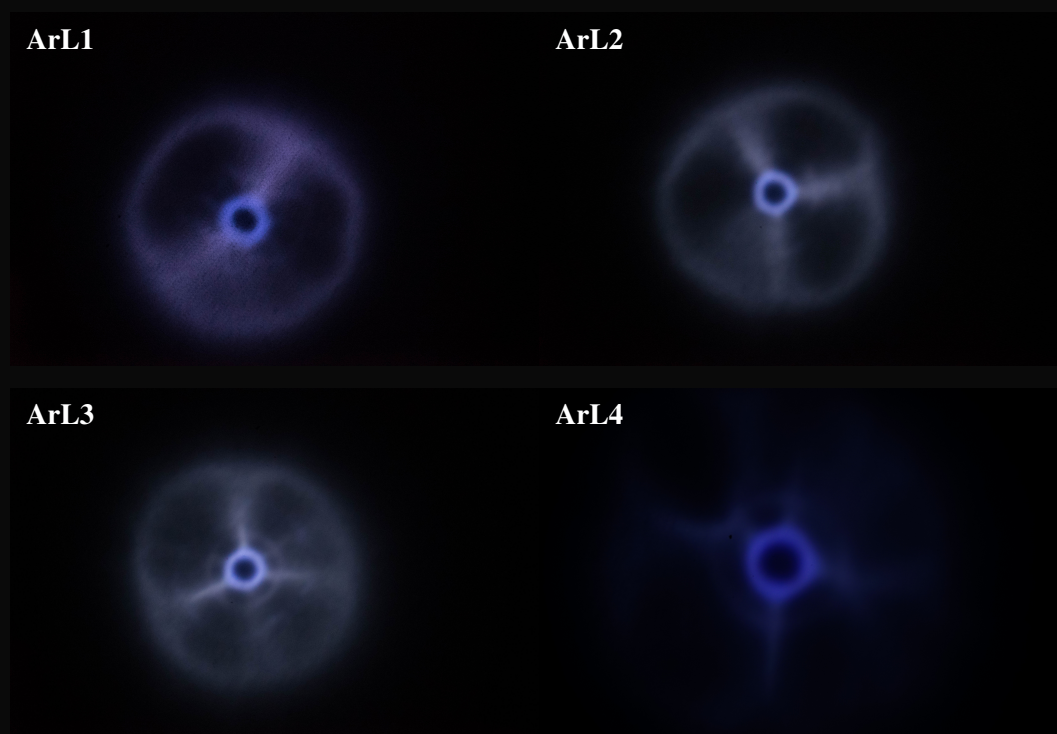


Argon

Quartz



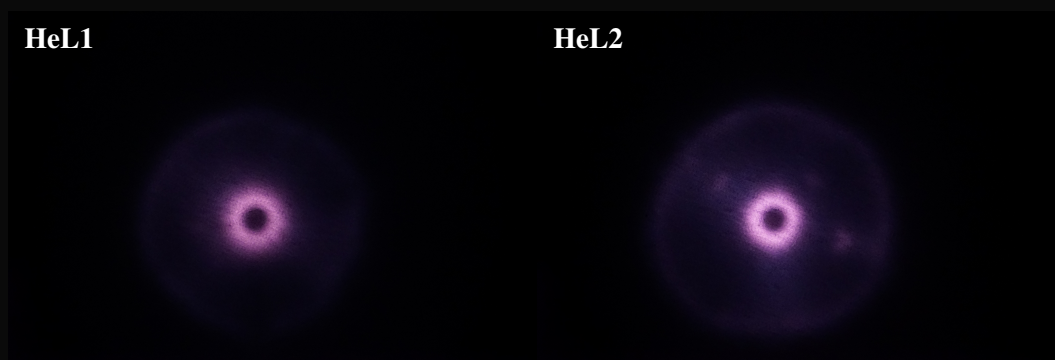
Lithium Niobate



**Helium
Quartz**



Lithium Niobate





*Appendix B***SUPPLEMENTARY MATERIAL: TOROIDAL PLASMOID
GENERATED THROUGH EXTREME HYDRODYNAMIC
SHEAR**

The following section was adapted for inclusion in this thesis with permission from

M. Gharib, S. Mendoza, M. Rosenfeld, M. Beizai, & F.J. Alves Pereira, Toroidal plasmoid generation via extreme hydrodynamic shear, Proc. Natl. Acad. Sci. U.S.A. 114 (48) 12657-12662, <https://doi.org/10.1073/pnas.1712717114> (2017).

B.1 SI Detailed Calculations

Electron Density

We define the current density as $j = n_e \cdot e \cdot v_d = E/\eta$, where n_e is the electron density, e the elementary charge, v_d the electron mean drift velocity, E the electric strength, and η the plasma resistivity. We have $\eta = m_e(\nu_{ei} + \nu_{en})/(n_e e^2)$, where m_e is the electron mass and ν_{ei} and ν_{en} are the electron–ion and electron–neutral collisional rates, respectively. Assuming electron–electron collisions are negligible, the momentum balance equation results in $\nu_{ei} = eE/(m_e v_d) - \nu_{en}$ at steady state.

The electron–neutral collision rate is $\nu_{en} = \sigma_e n_n v_{Te}$, where σ_e is the electron–neutral collisional cross-section, n_n the density of neutral gas particles, and v_{Te} the electron thermal velocity defined (Huba, 2004) as $v_{Te} = \sqrt{T_e/m_e} \approx 6.81 \times 10^7 \text{ cm s}^{-1}$ for $T_e = 2.64 \text{ eV}$. Using the average atomic radius of the two main constituents of air (nitrogen and oxygen), the collision cross-sectional area σ_e is $3.4 \times 10^{-16} \text{ cm}^2$. Finally, the electron drift velocity v_d is a function of the reduced field strength E/n_n (Rajasekaran, Bibinov, and Awakowicz, 2012). For the values measured with the air plasma, the mean electron drift velocity v_d is $3.06 \times 10^7 \text{ cm s}^{-1}$.

The Spitzer resistivity associated with electron–ion collisions is given (Bellan, 2008) by $\eta_s = Ze^2 \sqrt{m_e} 4\sqrt{2\pi} \ln \Lambda / [3(4\pi\epsilon_0)^2 T_e^{3/2}]$, where Z is the charge state, ϵ_0 the free space permittivity, and T_e the electron temperature in eV. $\ln \Lambda$ is the Coulomb logarithm, which is defined (Bellan, 2008) as $\ln \Lambda = 23 - \ln(n_e^{1/2}/T_e^{3/2})$ for $T_e < 10 \text{ eV}$. By substituting the term $m_e \nu_{ei}/(n_e e^2)$ in the definition of the total resistivity η by the Spitzer resistivity η_s , n_e can be determined for $T_e = 2.64 \text{ eV}$ and $Z = 1$, for we assume singly ionized air molecules in our calculations. We find $\ln \Lambda = 4.64$, a plasma resistivity $\eta = 12.6 \times 10^{-5} \Omega \text{ m}$, and an electron density $n_e = 1.62 \times 10^{17} \text{ cm}^{-3}$. Electron pressure is $6.84 \times 10^4 \text{ Pa}$. Ultimately the current density is $j \approx 7.93 \text{ mA } \mu\text{m}^{-2}$, corresponding to a dissipated power $j \cdot E \cdot V_p \approx 80 \text{ W}$ based on a plasma volume $V_p \sim 10^{-14} \text{ m}^3$ (see main-text plasma overview, panel B). To validate this estimate, we note that the power dissipated by a jet with diameter $85 \mu\text{m}$ flowing at 350 m s^{-1} and impacting on a wall is $\sim 120 \text{ W}$, assuming total dissipation on impact.

Plasma Oscillation

Plasma frequencies are given by the Tonks–Langmuir relations (Huba, 2004) $\omega_{pe} = \sqrt{n_e e^2 / (m_e \epsilon_0)}$ and $\omega_{pi} = \sqrt{n_i (Ze)^2 / (m_i \epsilon_0)}$, respectively the plasma–electron and the plasma–ion frequencies, with n_i being the ion density and $m_i = A \cdot m_p$ the ion

mass, where A is the atomic number and m_p the proton mass. Assuming singly ionized N_2 molecules such that $Z = 1$ and $A = 14$, and under the assumption of quasi-neutrality implying $n_i \sim n_e$, we obtain $\omega_{pi} = \omega_{pe} \sqrt{m_e/m_i}$. The ion field oscillates at a much lower frequency than the electron field as a consequence of the $\sim 25,000$ mass ratio between N_2^+ and the electron. For $n_e = 1.62 \times 10^{17} \text{ cm}^{-3}$, we obtain $\omega_{pe}/(2\pi) = 3.61 \text{ THz}$ and $\omega_{pi}/(2\pi) = 22.5 \text{ GHz}$.

Ion Acoustic Waves

In its simplest form, the dispersion relation for the ion acoustic wave is (Fenneman, Raether, and Yamada, 1973; Bellan, 2008) $\omega = kc_s/\sqrt{1 + k^2\lambda_{De}^2}$, where k is the wavenumber, c_s is the ion acoustic velocity given by $c_s = \omega_{pi}\lambda_{De} = \sqrt{T_e/m_i}$, and λ_{De} is the Debye length defined as $\lambda_{De} = \sqrt{\epsilon_0 T_e/n_e e^2}$, T_e being expressed in eV. We obtain $\lambda_{De} \approx 30 \text{ nm}$ and $c_s \approx 4.25 \times 10^5 \text{ cm s}^{-1}$. If $k^2\lambda_{De}^2 \gg 1$, ω saturates as expected at $c_s/\lambda_{De} = \omega_{pi}$, the plasma-ion frequency. If we now assume that $k^2\lambda_{De}^2 \ll 1$, the wavenumber matching the 36 MHz peak is $k = \omega/c_s \approx 5.32 \times 10^4 \text{ rad m}^{-1}$, leading to $k^2\lambda_{De}^2 \approx 2.55 \times 10^{-6}$, which validates the initial assumption.

Furthermore, the onset of ion acoustic instabilities occurs when $\omega_{pi}v_d/\nu_{in}v_{Te} \geq \sqrt{27/2\pi}$ (Fenneman, Raether, and Yamada, 1973), where $\nu_{in} = \sigma_i n_n v_{Ti}$ is the ion-neutral collision frequency, with $\sigma_i = 1.47 \times 10^{-15} \text{ cm}^2$ being the ion-neutral collisional cross-section and v_{Ti} the ion thermal velocity defined as $v_{Ti} = \sqrt{T_i/m_i}$ (Huba, 2004). Taking T_i as equal to the mean gas temperature $T_g = 1250 \text{ K} \approx 0.11 \text{ eV}$, we obtain $v_{Ti} = 8.6 \times 10^4 \text{ cm s}^{-1}$. Hence the ion-neutral collision frequency is $\nu_{in} = 3.6 \times 10^8 \text{ s}^{-1}$ and $\omega_{pi}v_d/\nu_{in}v_{Te} \sim 178$. The condition for the onset of ion acoustic instabilities is therefore also satisfied.

Water pH

In the air gas phase (Table B.1 A–H), N_2 and O_2 split through electron collisions with bond-dissociating energies of 941 kJ mol^{-1} and 498 kJ mol^{-1} , respectively. The highly reactive nitrogen atoms recombine into N_2^+ ions and free electrons through associative ionization. Combination with O_2 atoms and the Zeldovich reactions leads to nitric oxide $\cdot\text{NO}$ (Table B.1 C and D), which bonds with oxygen to form nitrogen dioxide $\cdot\text{NO}_2$. Highly soluble in water, $\cdot\text{NO}_2$ hydrolyses with H_2O molecules at the air–water interface to produce nitrous acid HNO_2 and nitric acid HNO_3 . In aqueous solution and because of the polar nature of water, these acids split into hydrons H^+ and the respective conjugate bases NO_2^- (nitrite) and NO_3^-

(nitrate). Hydrogen ions H^+ bond with H_2O to form the hydronium ion H_3O^+ , the conjugate acid of water. In the absence of plasma, nitrite-N in water sampled at the wafer edge (SI Materials and Methods) was not detected whereas nitrate-N levels averaged to 0.09 ppm. When the air corona was active, nitrite-N and nitrate-N levels averaged 0.025 ppm and 0.15 ppm, respectively, corresponding to a nitrite concentration of $1.78 \times 10^{-6} \text{ mol L}^{-1}$ and to an effective nitrate concentration of $2.49 \times 10^{-6} \text{ mol L}^{-1}$. The corresponding concentration of cations $\text{H}^+/\text{H}_3\text{O}^+$ added to the water is $4.28 \times 10^{-6} \text{ mol L}^{-1}$, causing the pH to drop from 5.6 to 5.17.

Solvated Electrons

The positive ions produced in air are accelerated by the electric field, bombarding H_2O molecules at the air–water interface to produce (Table B.1 I–O) excited water molecules H_2O^* , H_2O^+ cations, and hydrated electrons $e_{(aq)}$ (Herbert and Coons, 2017). Short-lived in water (Buxton et al., 1988; K. R. Sieferrmann et al., 2010) and confined within nanometers from the surface (Rumbach et al., 2015), these solvated electrons are released into the gas phase through a chemical tunneling effect (Hart and Anbar, 1970) (Table B.1 L–O). This secondary pathway to electron emission needs only 1.56 eV (Cserfalvi and Mezei, 1996). A low pH promotes the protonation of water and the reaction rate between H_3O^+ and the hydrated electrons, enhancing the electron release (Mezei and Cserfalvi, 2007).

Energy Balance

In air, dissociation of O_2 (9.75 eV) and N_2 (5.16 eV), excitation and ionization of air molecules ($> 2.64 \text{ eV}$), and secondary electron emission (1.56 eV) amount to $\sim 19.1 \text{ eV}$. With helium, H_2O dissociation (5.11 eV), excitation and ionization of OH ($> 4 \text{ eV}$) and H ($> 10.2 \text{ eV}$), and excitation of He atoms ($> 3.37 \text{ eV}$) amount to $\sim 22.7 \text{ eV}$.

B.2 SI Materials and Methods

General Information

The experimental pump setup is based on a DSF-model Haskell fluid pump driven by building air pressure. The air pressure delivered to the system was kept below 159 psi, resulting in a maximal liquid output pressure of 16,500 psi and a corresponding jet speed of 430 m s^{-1} . A water reservoir delivers a constant, deionized water supply through the pump to an AccuStream waterjet system equipped with a $100 \mu\text{m}$ diameter ruby nozzle. The jet contracts due to the vena contracta effect to a

diameter of 85 μm and keeps this value until the point of impingement. The distance between the nozzle tip and the surface was kept at 20 mm to avoid jet breakup and spray formation. The water delivered to the system was refreshed at least once daily to keep ion concentration low in the system. Water pH was measured with a Spectrum Technologies IQ 150 pH meter and water resistivity was measured with a Fluke 287 multimeter with dual brass probes.

Target wafers tested were methyl acrylate, niobium, lithium niobate (X-cut, Y-cut, Z-cut, polished, and fine ground), fused quartz, seeded quartz (polished and fine ground), borosilicate glass, silicon, and sapphire (polished and fine ground). Around the target wafers, a hermetic acrylic container was designed that allowed gas input for gas exchange, a positive pressure drain for water and air purging, and a cutout beneath the wafer stage, which allowed optical access through the wafers to the impinging jet site.

A Nikon D90 or a Sony A7S digital camera was adapted to fixed-focus microscope objectives for imaging through the optical cutout, with constant settings and neutral white balance used for all images unless otherwise noted. A Quest U BRC112P-U CCD array spectrometer (B&W Tek, Inc.) was used to record optical spectra in the range of 300–850 nm through the same optics and was equipped with a 10 μm slit giving a spectral resolution of 0.3–0.4 nm. RF as well as electric field measurements were done digitally with a Tektronix MDO3024 mixed-domain oscilloscope.

Calibration of Jet Velocity

The digitally measured jet diameter at the impingement distance in combination with the average liquid flow rate through the jet nozzle allowed the back calculation of the average velocity of the jet at a range of input pump pressures. To measure the flow output, the jet was turned on for 30 s while the output water was captured in a graduated cylinder. Input pressures from 30–150 psi were tested, resulting in an output volume ranging from 27.2–75.2 ml during this period of 30 s. Given a jet diameter of 85 μm , a quadratic regression of the measurements provided the following approximation for the jet velocity U ,

$$U(P) = -0.0255P^2 + 6.29595P - 3.07143 \quad (R^2 = 0.998), \quad (\text{B.1})$$

where U is given in m s^{-1} and P is the pump pressure given in psi. The maximum tenable average jet velocity was 428 m s^{-1} .

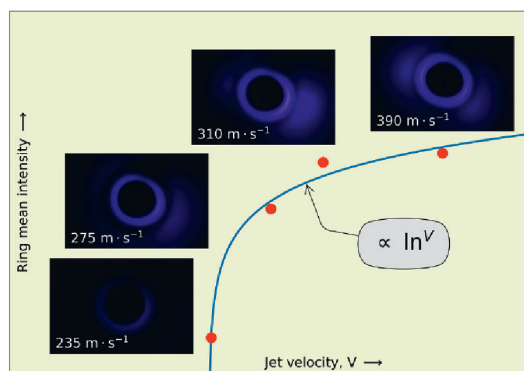


Figure B.1: Increase in corona luminosity with increase in impingement velocity. As the water pressure rises, the outlet velocity increases and a corresponding monotonic increase in light intensity follows. The luminescent patterns were observed on a polished quartz wafer. We noted that the intensity of the luminescent ring increased logarithmically with the velocity. Photographs were taken at ISO 1,000 and a 1/5 s exposure time with a Nikon D90 camera and a 50× objective lens.

Target Materials

We observed a variety of behaviors from the dielectric materials tested. Methyl acrylate was quickly damaged by the jet, yet did generate luminescence. Borosilicate glass generated luminescence and cracked after several minutes, releasing glass shards. Niobium and silicon wafers were durable but failed to generate any luminescence. Quartz, lithium niobate, and sapphire samples generated luminescence and were hard enough to withstand the pressure of the impinging jet. Of these latter materials, single crystal quartz (SiO_2) and single crystal lithium niobate (Z-cut LiNbO_3) required significantly lower activation water pressures than did sapphire. With an optical transmittance higher than 50% up from wavelengths of ~ 150 nm for SiO_2 and ~ 320 nm for LiNbO_3 , these materials were also suitable for optical and spectral analysis. Surface quality was either polished (microroughness $Ra < 10 \text{ \AA}$) or fine ground (microroughness $Ra < 1 \text{ }\mu\text{m}$).

Water Resistivity

Water resistivity in the reservoir measured an average of $18 \text{ M}\Omega \text{ cm}$. Failure to refresh water stores for a period of 18 h resulted in a drop in resistivity from $18 \text{ M}\Omega \text{ cm}$ to $4.5 \text{ M}\Omega \text{ cm}$. At $4.5 \text{ M}\Omega \text{ cm}$, the corona was not stable, intermittent, or at times absent.

Measurement of pH

To assess any pH change in the deionized water that was exclusively a result of the luminescent process itself, water was collected into a glass reservoir following jet impingement in the presence and in the absence of a corona. The pH meter was calibrated with stock buffer solutions. Source water pH was measured to be 7.1 on average. Running the water through the pump tubing lowered the pH to 5.6 ± 0.03 at the nozzle.

Physico-Chemical Characterization of Water

We tested the presence of nitrites and nitrates in the water upstream and downstream of the impinging site, in the absence and presence of air corona, and when the gas was changed from air to helium. The water was sampled at the jet outlet before impingement and at the edge of the wafer plate after impingement. Nitrite levels were measured with a nitrite-nitrogen (NO_2^- -N) Ultra Low Range HI764 photometer test kit from Hanna Instruments. The NO_2^- -N detection method uses an adaptation of the EPA diazotization method 354.1. The effective range of the photometer kit is from 0 ppb to 200 ppb NO_2^- -N ± 10 ppb $\pm 4\%$ of reading at 25 °C. Resolution is 1 ppb. Nitrate levels were measured with a low-range nitrate-nitrogen (NO_3^- -N) photometer test kit from Environmental Express. The NO_3^- -N detection method uses eukaryotic nitrate reductase to convert available nitrates into nitrites, followed by the same diazotization method used to measure nitrite-nitrogen levels. The effective range of the photometer kit is from 0.05 ppm to 2.0 ppm NO_3^- -N $\pm 10\%$. Total measured NO_2^- -N results from both actual NO_2^- -N and converted NO_3^- -N. Therefore, the actual content of nitrates is calculated using the separate measurement of the concentration of nitrites alone with the nitrite-nitrogen kit.

Imaging

Although light emitted by the luminescent phenomenon was detectable without magnification, viewing the corona through the optical cutout with high magnifications revealed important topological features. Fig. B.4 was taken at ISO 640 with a 1/5 s exposure time, using a Nikon D90 camera mounted on a 160 mm-long microscope tube coupled with a 50 $\times$ objective lens. Using a Sony A7S camera with a 1/60 s exposure time and a 10 $\times$ objective lens, photographs in Fig. 1.1C, Fig. B.4, and Fig. B.5 were captured at ISO 20,000, whereas photographs in Fig. 1.2A, Fig. B.6, and Fig. B.7 were taken at ISO 12,800.

Optical Emission Spectroscopy

To calibrate the sensitivity of the B&W Tek, Inc. spectrometer across as much of its range as possible, optical spectra of an Oriel sun simulator (LCS-100 Solar Simulator, model 91104A) were recorded without magnification. True spectral data of the same Oriel sun simulator were provided by the manufacturer, which were then compared with the readout of our spectrometer. A built-in calibration function adjusted values accordingly. In the experimental setup, the optical fiber from the spectrometer was connected to a full-pass collimator with C-mount threading. This adapter allowed the Nikon D90/Sony A7S cameras and the spectrometer collimator to be interchanged, using the same objective lens without moving it. Connecting both devices to the same objective allowed all light passed to the camera sensor to also pass to the spectrometer CCD. The focal tube connected to the microscope objectives was mounted beneath the acrylic container and the target wafer. Before each spectroscopy session, all sources of ambient light were shut off to ensure sterility in the readings. A background signal was recorded and stored each time. For both background and signal spectra recordings, the integration time was set to 200 ms with 15 time averages and a multiplier of 10 for a total recording time of 30 s. Dark subtraction and intensity calibration were also implemented for all spectra.

RF Measurements

Using the spectrum analyzer function of the Tektronix digital oscilloscope, RF emissions of the luminescent phenomenon were recorded between 1 MHz and 100 MHz for lithium niobate and seeded quartz wafers. 22AWG enamel-coated wire was wound around an acrylic cylinder 10 mm in diameter and glued with thermoplastic adhesive to maintain its shape. The acrylic core was then removed to leave the structure forming the antenna as shown in Fig. B.2. RF emissions were recorded and averaged over 512 samples (≈ 15 s) with the antenna firm in its measuring position. Before jet initialization, background emissions were also recorded.

Electrical Grounding of Wafer

A brass needle was positioned underneath the wafer on a three-axis microtraverse system, which allowed one to press the needle against the plate and move it radially away from the point of impingement on the reverse side of the wafer. The bent tip of the needle allowed some force to be applied to the needle to ensure contact. The opposite end of the needle was then connected to an electrical ground.



Figure B.2: RF antenna. The customized RF antenna is shown in its setup position relative to the jet nozzle. The antenna axis is collinear with the jet axis.

Electric Field Mapping

Probing the interior of a plasma is usually done by sweeping the voltage on the probe and measuring the resultant probe current. Our analysis, however, was not for the interior of the plasma but for the electric field emitted by it. Measuring equipotential surfaces and mapping electric field lines are commonly done with a probe at a fixed potential paired with a floating probe which measures the potential at various points in the electric field. The potential of the interior of the plasma was determined to be consistent enough to act as a reference potential. The electric field exterior to the plasma was then measured with a free probe at various points throughout the field. Mapping the electric field via voltage measurements was done using the digital voltmeter function on the Tektronix oscilloscope. Two brass needles were encased with a two-coat hydrophobic and electrically insulating layer. The tips were then cleaned of this layer to expose a small conductive area. Each needle was then connected to different three-axis microtraverses. The reference probe tip was positioned within $75\ \mu\text{m}$ of the jet boundary. When the probe tip was moved closer, the position of the probe interfered with the jet structure. The other probe tip was initially positioned adjacent to the reference probe and then moved incrementally outward along the horizontal and vertical axes, independently. Voltage measurements were recorded and averaged for 30 s at each point. A contour

map of the averaged values of voltages around the site of impingement is shown in the main-text potential-field map. The probes were set up such that the positive voltmeter terminal remained near the jet annulus as the reference electrode. In this configuration all measured values were negative, indicating a surplus of negative charge carriers (electrons) around the jet annulus compared with the charges in the air at distances away from the corona. To determine the electric field lines, the sparse data of the potential field were interpolated using Hermite polynomials and spatially derived between all data points, resulting in the white arrowed lines shown in the main-text potential-field map. In that potential-field map, extrapolated data are reported as dashed black lines in the region between $75\text{ }\mu\text{m}$ and the jet boundary.

Jet Flow Simulation

The water jet of diameter $85\text{ }\mu\text{m}$ impinged vertically on a solid wall with a velocity of 250 m s^{-1} , corresponding to a Reynolds number of 21,250 based on the jet diameter. The domain of computation was $240\text{ }\mu\text{m} \times 1,000\text{ }\mu\text{m}$ in the axial and radial directions, respectively. Zero velocity was imposed at the wall and zero-gauge pressure was specified at the outer boundaries. The 2D, laminar, incompressible, axisymmetric, unsteady two-phase flow Navier–Stokes equations were solved until steady state was reached. The implicit volume-of-fluid method together with the compressive interface-capturing scheme for arbitrary meshes was used to model the sharp interface between the water jet and the surrounding air. The Ansys Fluent 16.0 CFD package was used to solve the discrete problem. The second-order upwind spatial scheme with the PRESTO! pressure approximation was used. Following a mesh-independence test, the 2D quadrilateral mesh consisted of 180,000 nodes with four-level mesh adaptation in the vicinity of the wall and of the free surface. The node spacing near the wall and the free surface was below $0.013\text{ }\mu\text{m}$ and $0.2\text{ }\mu\text{m}$, respectively.

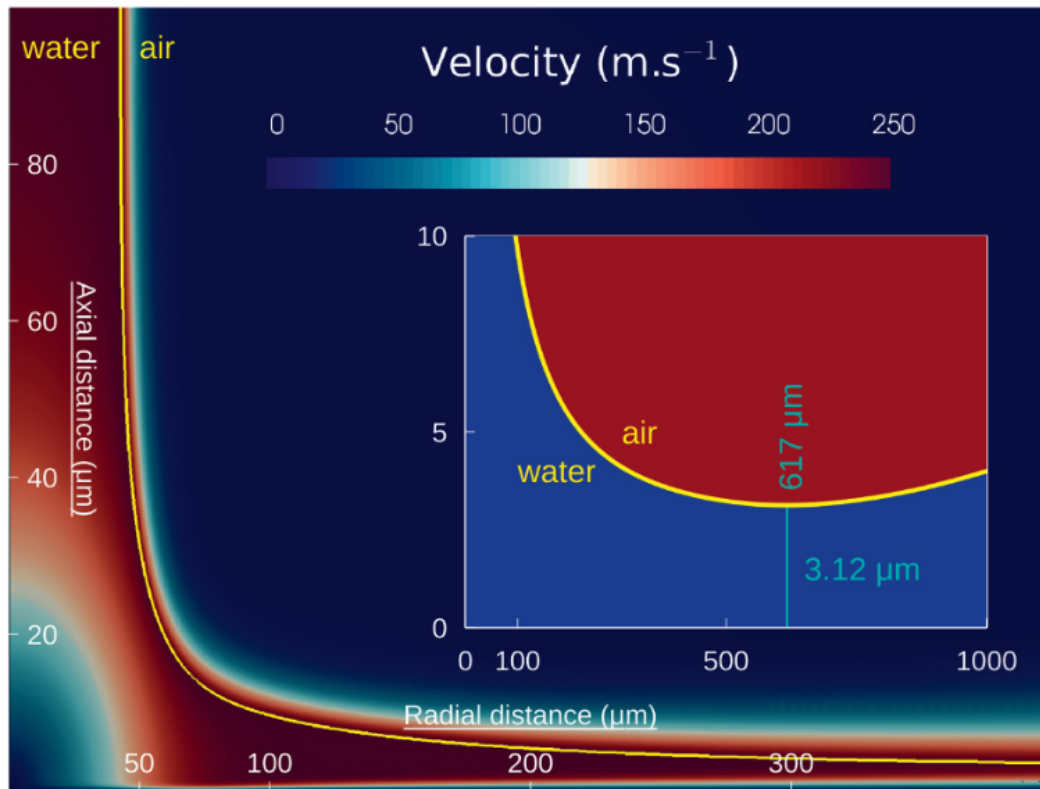


Figure B.3: Computational fluid dynamics simulation of the impinging jet flow. The 2D calculations were done for a nominal jet speed of 250 m s^{-1} . The color plot shows the map of the velocity magnitude, with the yellow line representing the air–water interface. Inset outlines the thinning of the wall jet to a minimum of $3.12 \mu\text{m}$ at a radial distance of $617 \mu\text{m}$ from the jet axis. Scales of axes are in micrometers.

B.3 Gallery

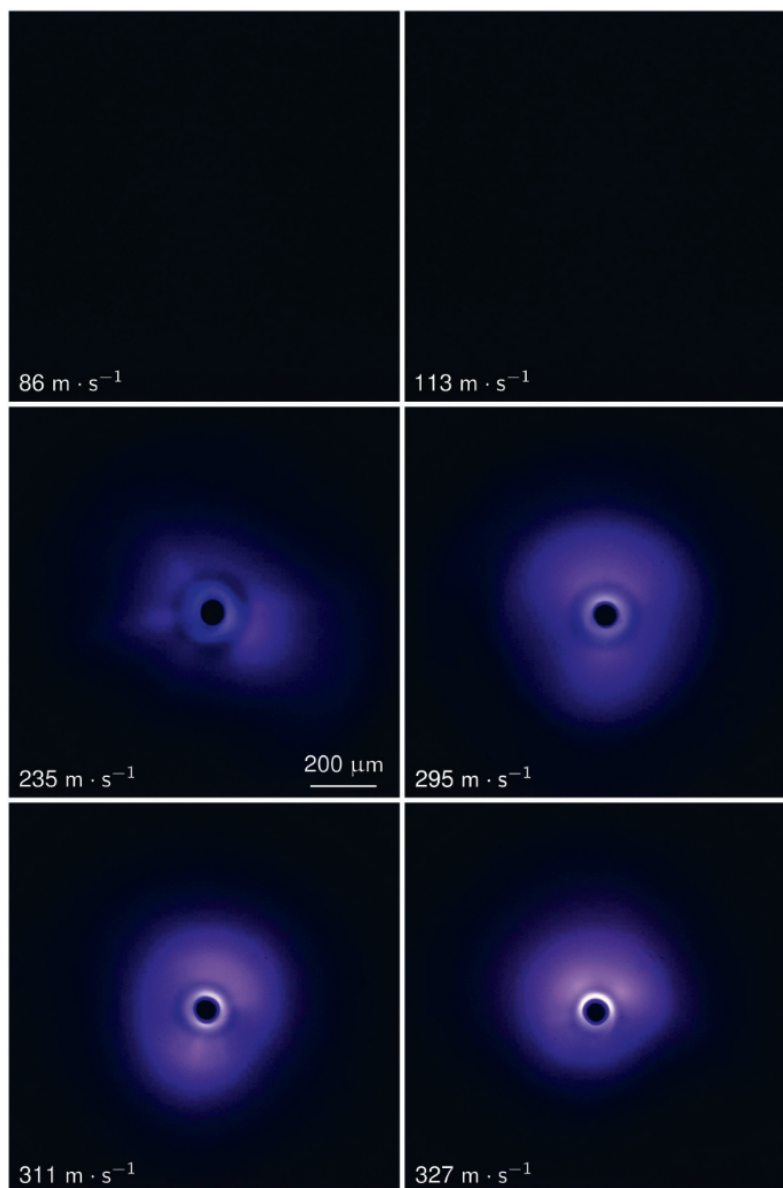


Figure B.4: Luminescence in air on a polished quartz surface. Jet velocity is varied from $86\text{--}327 \text{ m} \cdot \text{s}^{-1}$. Photographs were taken at ISO 20,000 and a $1/60 \text{ s}$ exposure time with a Sony A7S camera and a $10\times$ objective lens. Color levels of images have been enhanced by 33% for better display.

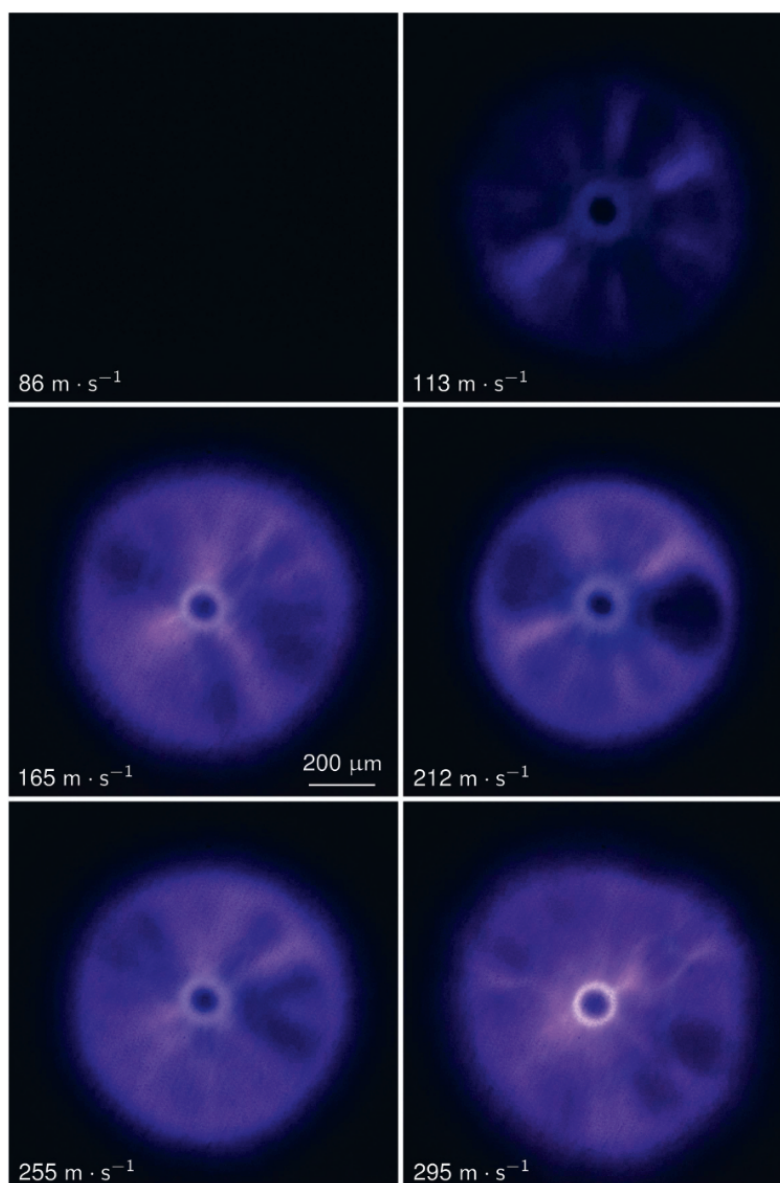


Figure B.5: Corona in air on a fine-ground lithium niobate surface. Jet velocity is varied from 86–295 m s⁻¹. Photographs were taken at ISO 20,000 and a 1/60 s exposure time with a Sony A7S camera and a 10× objective lens. Color levels of images have been enhanced by 33% for better display.

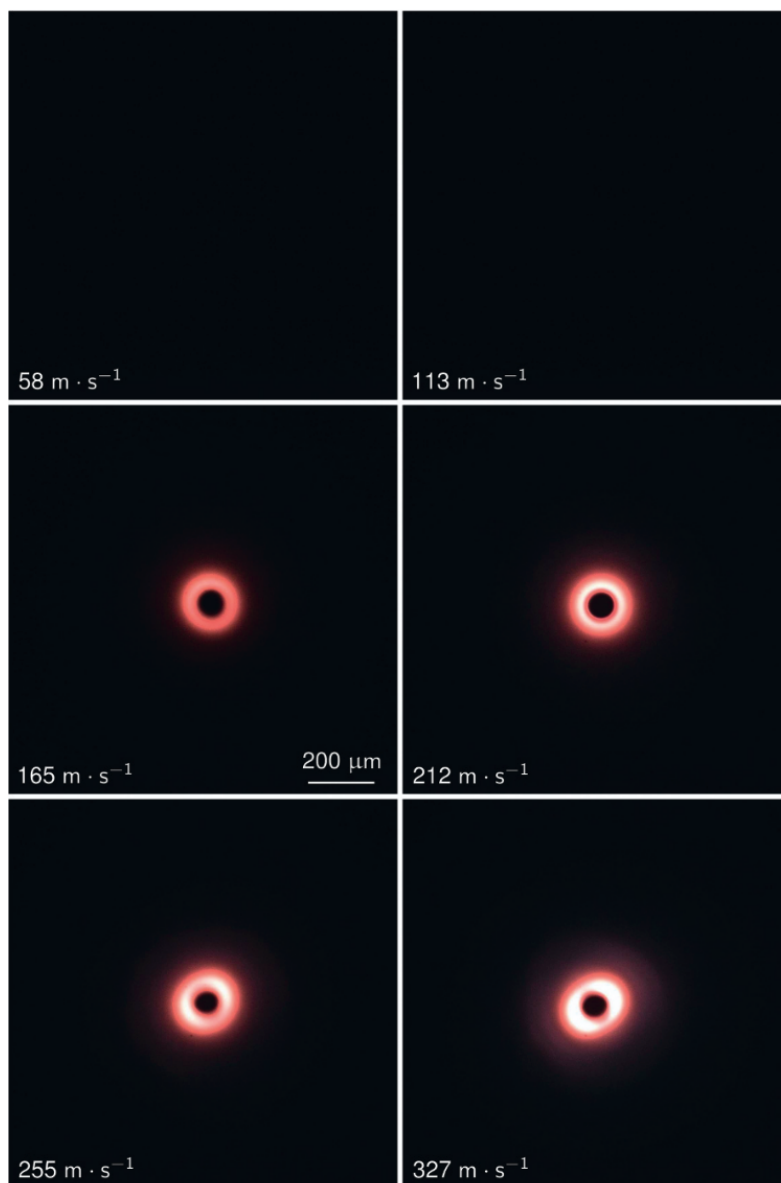


Figure B.6: Luminescence in helium on a polished quartz surface. Jet velocity is varied from 58–327 $\text{m} \cdot \text{s}^{-1}$. Photographs were taken at ISO 12,800 and a 1/60 s exposure time with a Sony A7S camera and a 10 $\times$ objective lens. Color levels of images have been enhanced by 33% for better display.

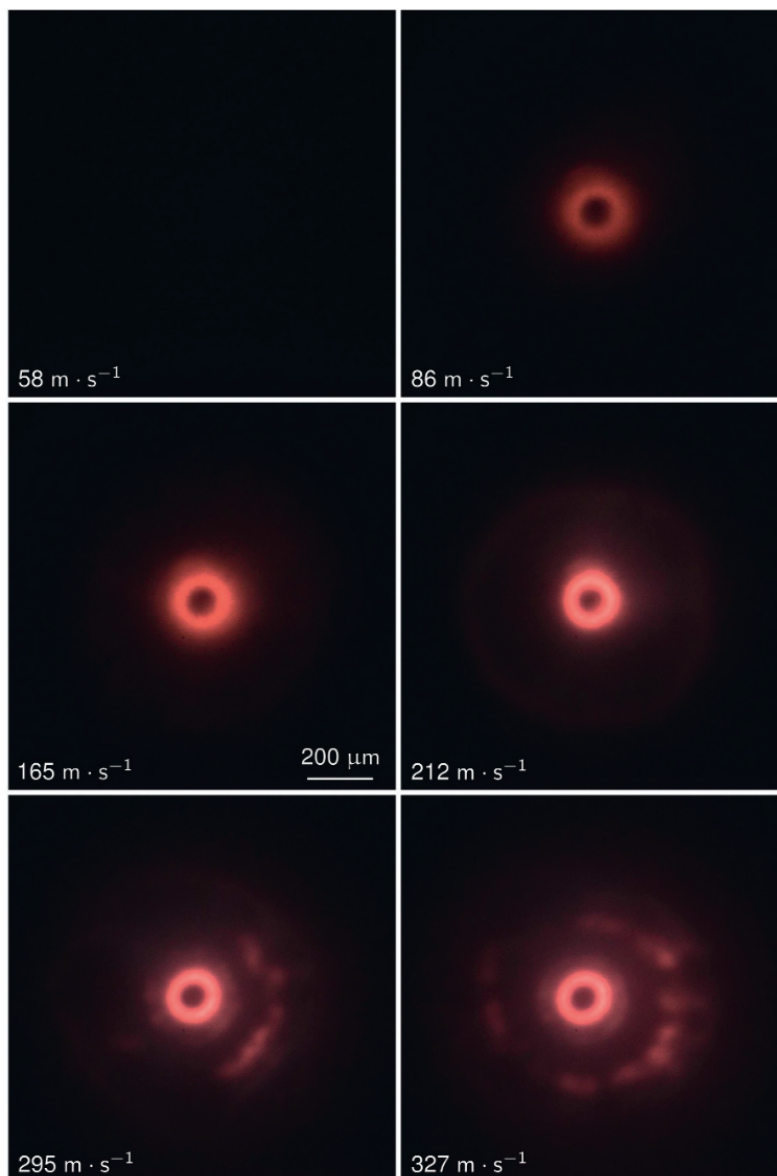


Figure B.7: Luminescence in helium on a fine-ground lithium niobate surface. Jet velocity is varied from 58–327 $\text{m} \cdot \text{s}^{-1}$. Photographs were taken at ISO 12,800 and a 1/60 s exposure time with a Sony A7S camera and a 10× objective lens. Color levels of images have been enhanced by 33% for better display.

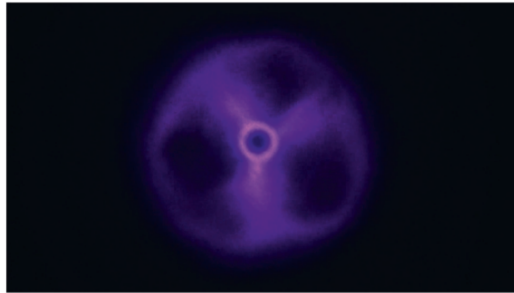


Figure B.8: Movie S1 Luminescence in air on a rough LiNbO_3 surface. The movie was taken with a $10\times$ optical magnification and shows how the luminescence pattern is affected by the surface roughness. We can observe the formation of radial streamers that bridge the central toroidal plasma with a plasma ring located approximately at $500\text{ }\mu\text{m}$ from the jet impinging site. The streamers are seen to interact collectively and to give a visual perception of rotation, resembling the streamers seen in plasma balls.



Figure B.9: Movie S2 Effect of high electric potential on the plasma stability and viability. The movie was taken from the impinging side and shows that the application of a 50 kV potential between the wafer and the nozzle, either positive or negative, leaves the plasma structure unaffected. The luminescence was generated in a mixed atmosphere of air and helium. Secondary streamers are seen that bridge the nozzle to the wafer away from the luminescent structure at the jet impingement site. They are not connected and do not interfere with this latter phenomenon.

Table B.1: Possible chemical reactions in air, in liquid water, and at the interface for the air plasma (corona) and helium plasma

Reaction identification letter	Reaction type or product	Chemical equation
<i>In air with corona</i>		
A)	N ₂ /O ₂ dissociation	$N_{2(g)} + e^* \Rightarrow 2N_{(g)} + e$ $O_{2(g)} + e^* \Rightarrow 2O_{(g)} + e$
B)	Associative ionization	$N_{(g)}^* + N_{(g)} \Rightarrow N_{2(g)}^+ + e$
C)	Nitric oxide	$2N_{(g)} + 2O_{(g)} \Rightarrow 2 \cdot NO_{(g)}$
D)	Zeldovich exchange reactions	$N_{2(g)} + O_{(g)} \Rightarrow \cdot NO_{(g)} + N_{(g)}$ $N_{(g)} + O_{2(g)} \Rightarrow \cdot NO_{(g)} + O_{(g)}$ $2 \cdot NO_{(g)} + O_{2(g)} \Rightarrow 2 \cdot NO_{2(g)}$
E)	Nitrogen dioxide	
F)	Nitrous/nitric acids	$2 \cdot NO_{2(g)} + H_2O_{(l)} \Rightarrow HNO_{2(aq)} + HNO_{3(aq)}$
G)	Hydrons, nitrites, and nitrates	$HNO_{2(aq)} \Leftrightarrow H_{(aq)}^+ + NO_{2(aq)}^-$ $HNO_{3(aq)} \Leftrightarrow H_{(aq)}^+ + NO_{3(aq)}^-$
H)	Hydronium	$H_{(aq)}^+ + H_2O_{(l)} \Rightarrow H_3O_{(aq)}^+$
<i>At air–water interface with corona</i>		
I)	Impact ionization by positive ions ($\oplus$)	$\oplus + H_2O_{(l)} \Rightarrow H_2O^*, H_2O_{(aq)}^+ + e_{(aq)}$
J)	Dissociation	$H_2O_{(aq)}^+ \Rightarrow \cdot OH_{(aq)} + H_{(aq)}^+$
K)	Protonation	$H_{(aq)}^+ + H_2O_{(l)} \Leftrightarrow H_3O_{(aq)}^+$
L)	Recombination	$H_3O_{(aq)}^+ + e_{(aq)} \Rightarrow H_2O_{(l)} + H_{(aq)}$
M)	Diffusion	$H_{(aq)} \Rightarrow H_{(g)}$
N)	Ionization	$H_{(g)} + e^* \Rightarrow H_{(g)}^+ + e^* + e$
O)	Back diffusion and hydration	$H_{(g)}^+ + H_2O_{(l)} \Rightarrow H_3O_{(aq)}^+$
<i>In liquid water</i>		
P)	Dissociation	$H_2O_{(l)} + e^* \Rightarrow \cdot OH_{(aq)} + \cdot H_{(aq)} + e$ $H_2O_{(l)} + e^* \Rightarrow OH_{(aq)}^- + H_{(aq)}^+ + e$
Q)	Protonation	$H_{(aq)}^+ + H_2O_{(l)} \Leftrightarrow H_3O_{(aq)}^+$
R)	Self-ionization	$2H_2O_{(l)} \Leftrightarrow H_3O_{(aq)}^+ + OH_{(aq)}^-$
S)	Recombination	$H_3O_{(aq)}^+ + OH_{(aq)}^- \Leftrightarrow 2H_2O_{(l)}$

Appendix C

SUPPLEMENTARY MATERIAL: BREMSSTRAHLUNG RADIATION FROM A TOROIDAL PLASMA GENERATED THROUGH HYDRODYNAMIC SHEAR

The following section was adapted for inclusion in this thesis with permission from

Mendoza, S., Gharib, M. *Bremsstrahlung radiation from toroidal plasmas generated through hydrodynamic shear*. Sci Rep 15, 10494 (2025). <https://doi.org/10.1038/s41598-025-88250-7>.

Affiliation: Graduate Aerospace Laboratories, California Institute of Technology, Pasadena, CA 91125

C.1 Experimental Setup

Plasma Generation via Extreme Pressure Water-jet

A Haskel air-driven liquid pump (ASFD-60) is used to pump distilled water through a mounted ruby orifice (AccuStream PN1-23587-004, $\varnothing = 100 \mu\text{m}$). A coherent, collimated water jet exits the nozzle at a velocity of 200 m s^{-1} , corresponding to air and liquid drive pressures of 60 psi and 3600 psi, respectively. The exit velocity is modulated between approximately $150\text{--}300 \text{ m s}^{-1}$ by adjusting the drive pressure between roughly 20–120 psi.

The dielectric target employed in these experiments is Z-cut optical-grade lithium niobate (Precision MicroOptics POLN311311), positioned 1 cm from the orifice. The pump can be continuously operated, producing a persistent plasma in the gas due to triboelectrification of the hydrodynamic shear layer at the site of impingement.

The jet nozzle and target are enclosed within a hermetic quartz chamber with a regulated inlet for argon (Airgas UHP 99.999%) and regulated outlets for gas evacuation and water drainage. The lithium niobate plate is backed by a quartz optical window to provide optical access.

Imaging and Spectroscopy

Optical emission spectroscopy was performed using a B&W Tek spectrometer (BRC112EU) calibrated *in situ* with a NIST-traceable source lamp (Oriel 45W Halogen, 63358) for the 400–900 nm range. Light from the plasma was directly captured into the fiber-coupled spectrometer for quantitative measurements and directed with mirrors between the spectrometer and camera for qualitative imaging to allow simultaneous measurements.

A Sony A7S camera was used for all imaging. Qualitative imaging utilized a Laowa ultra-macro lens (VE2528C), while spectral calibration required an Olympus semi-apochromatic objective (LUCPLFLN20X). All photos were taken as raw images at the camera's native ISO (ISO 100).

For multispectral imaging, green and red optical bands were defined as $B_g = \{\lambda \mid 500 \text{ nm} \leq \lambda \leq 515 \text{ nm}\}$ and $B_r = \{\lambda \mid 618 \text{ nm} \leq \lambda \leq 632 \text{ nm}\}$, matching the set of chosen interference filters (Thorlabs FBH515-10 and Thorlabs FLH633-5). Due to the decreased light intensity from filtering to these bands, an exposure time of 4 seconds was used for multispectral imaging.

For numerical processing of multispectral image data, the raw images were demosaiced in software to extract the blue and red channels' data. These channels were chosen to represent the green-light and red-light measurements, respectively, as the camera's filter and sensor characteristics are most similar between the blue and red channels at the chosen wavelengths.

Optical Emission Spectrum

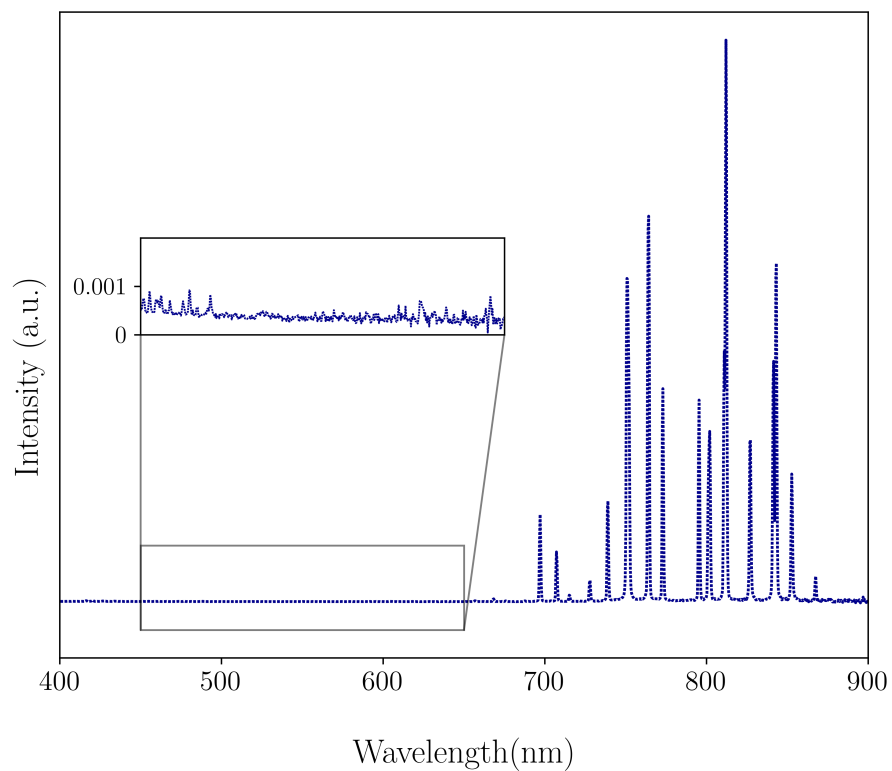


Figure C.1: Optical emission spectrum captured from the toroidal argon plasma from 400–900 nm. The inset magnifies the indicated segment of the visible spectrum (450–650 nm), showing a broadband continuum.

Spectral Error Analysis

To estimate the spectral error resulting from detector noise and spectral peaks in the chosen optical bands, the relative contribution of narrow spectral features was compared to broad spectral features. Within these bands, peak widths appear to be typically less than 1 nm, and are nearly indistinguishable from the detector noise. Fig. C.2 and Fig. C.4 show the FFT power spectrum (top) of spectral features comprising the total emissivity within the red and green band signals, respectively.

Since the continuum of interest spans many hundreds of nm, only the widest features correspond to continuum emission, while narrower features may correspond to spectral lines, as well as random noise. In the green band, this estimates the continuum contribution to be $\sim 1\%$, while in the red band, this contribution is $\sim 2.7\%$. Since the spectral contribution is primarily additive, while non-continuum FFT components assume zero-mean, the estimated contributed errors are maximally $\sim 2\%$ and $\sim 5.4\%$, respectively. Since these features cannot with complete certainty be ascribed to noise or spectral peaks, we interpret these percent contributions as the maximum error in the spectral band measurements.

Additionally, dark scans and raw data from the spectrometer are shown in Fig. C.3 for the red band, and in Fig. C.5 for the green band. These data clearly illustrate the relatively large dark signal resulting from long integration times, and that, among the narrow spectral features, a significant portion of the signal is attributable to dark noise.

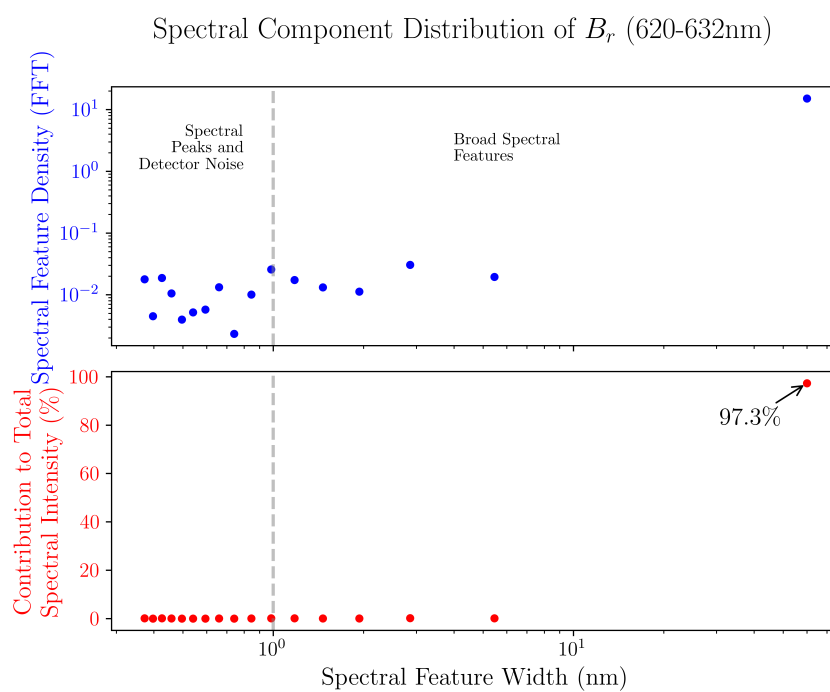


Figure C.2: The density of spectral features in the red band (top) is shown vs the feature width (in nm), computed via FFT. The percent contribution of each feature width to the total emissivity is shown in the bottom panel. The widest features (including “DC” components) correspond to continuum emission.

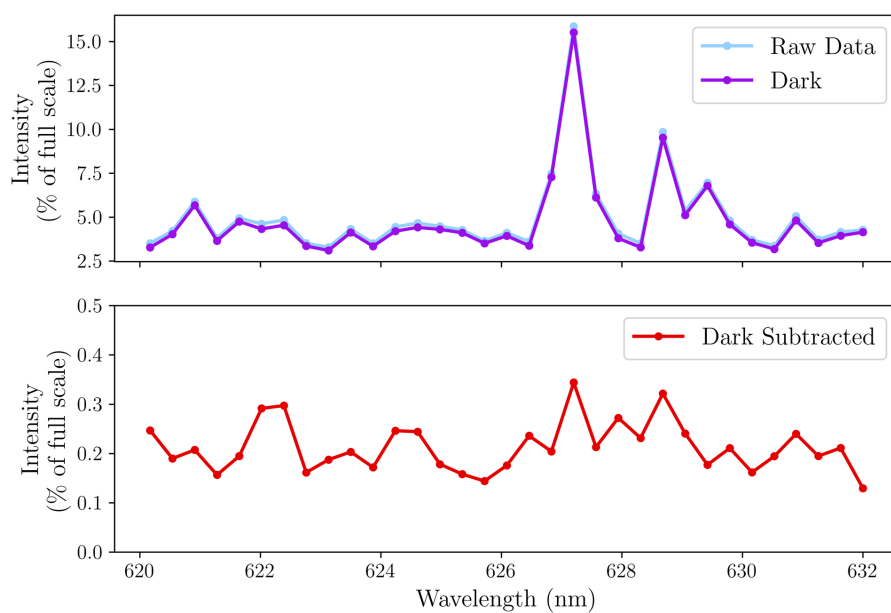
Spectrometer Raw and Dark Characteristics (B_r 620-632nm)

Figure C.3: Dark scan and raw data (top), and dark-subtracted data (bottom) are shown as percentage of maximum signal (16bit integer).

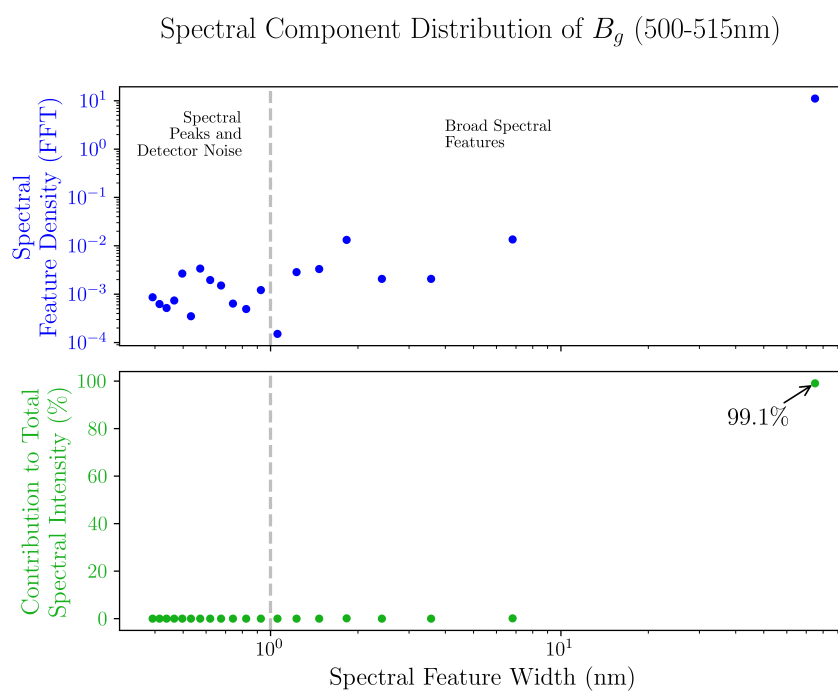


Figure C.4: The density of spectral features in the green band (top) is shown vs the feature width (in nm), computed via FFT. The percent contribution of each feature width to the total emissivity is shown in the bottom panel. The widest features (including “DC” components) correspond to continuum emission.

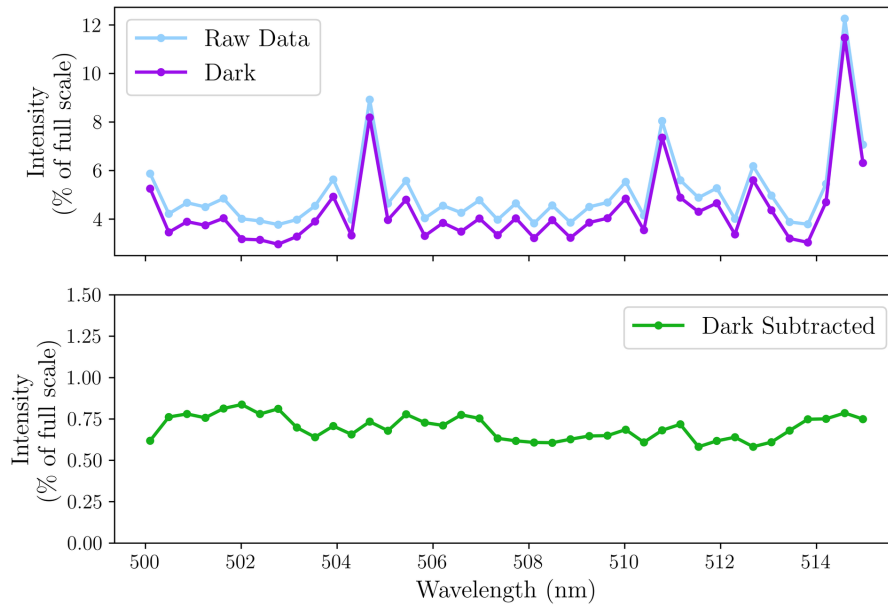
Spectrometer Raw and Dark Characteristics (B_g 500-515nm)

Figure C.5: Dark scan and raw data (top), and dark-subtracted data (bottom) are shown as percentage of maximum signal (16bit integer).

Plasma Thickness

Due to emissivity measurements being integrated over line-of-sight, the emissivity was normalized to an estimate of the plasma's thickness when calculating electron density. The thickness function used is shown in Fig. C.6.

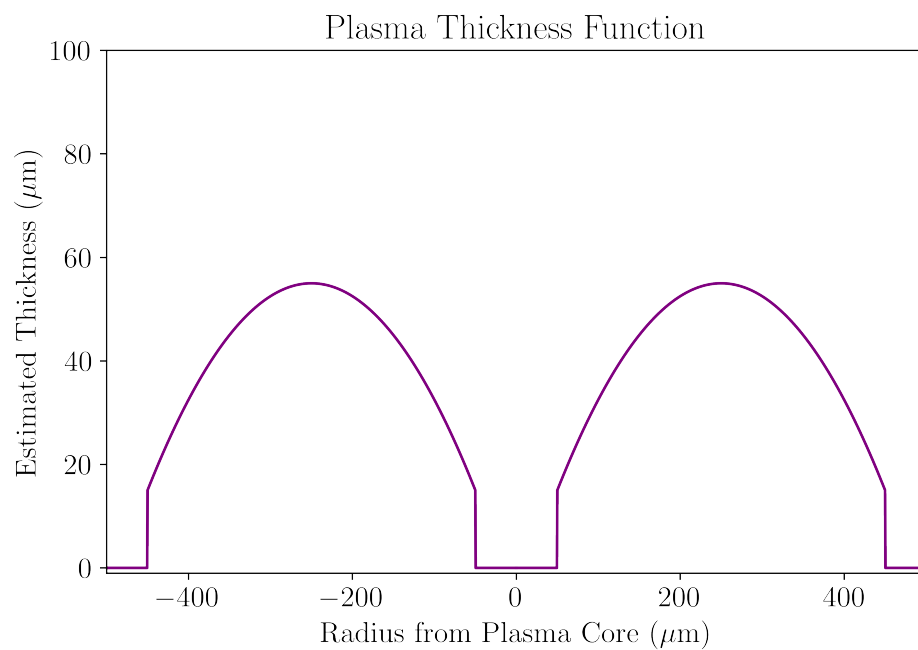


Figure C.6: Plasma thickness function used to calculate emissivity per thickness. Thickness function was generated based on approximate measurements of the cathode, anode, and plasma body.

Image Processing

For multispectral image analysis, the green and red channel images were aligned using a generic alignment algorithm to the greyscale images. Fig. C.7 shows an array of images resulting from different x- and y-shift combinations, with the central image (bordered in red) representing the T_e results of final alignment chosen to best represent the plasma images.

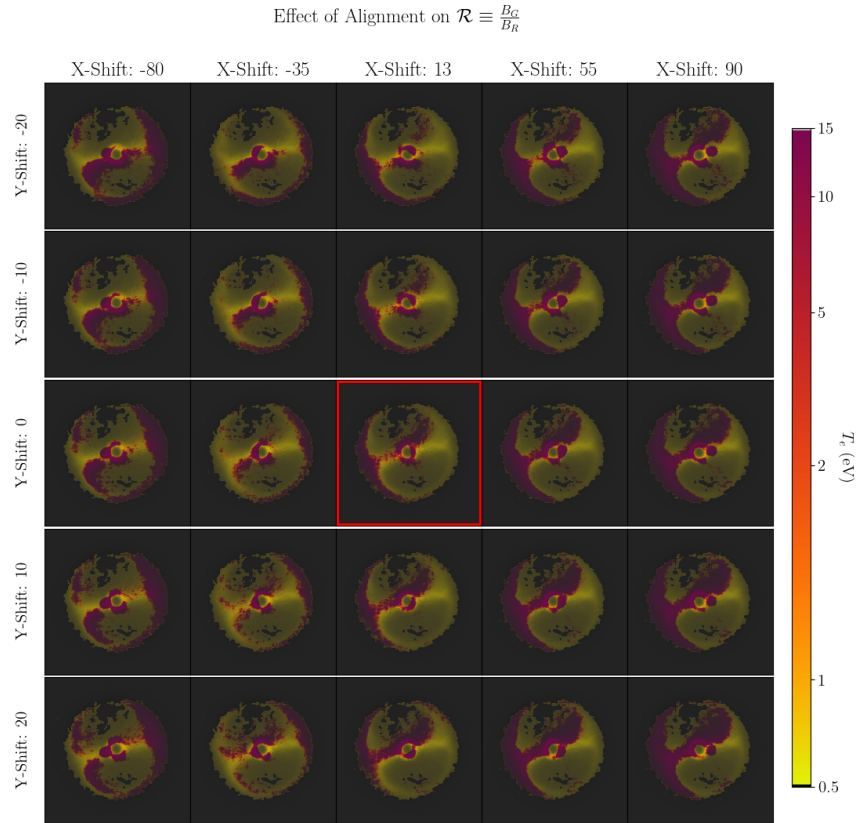


Figure C.7: Array of images showing estimated T_e distribution resulting from various x-shifts and y-shifts applied during the alignment process. The central image (bordered in red) represents the best alignment of the jet core and inner toroidal ring within both images.

Imaging Error Analysis

Independent from the spectroscopic data, camera sensor error is present in the image data used for these calculations. Under these imaging conditions, a summary of the contributions to the total imaging error is shown in Table C.1. For final cal-

ulation, images were downsampled using the indicated kernels to reduce standard measurement error.

Parameter	Background	Bromine	Unit
Read Noise ^a	11.2	11.2	electrons
Dark Error ^b	2.8	2.8	count
Spatial Error	2.9	2.9	count
Measurement Error ^c	6(7.5)	6(1.7)	count (%)
Averaging Kernel	9x9	3x3	pixels
Mean Standard Error	0.8	0.6	%
Total Error^d	5.5	2.1	%

Table C.1: Error analysis of camera sensor measurements. ^aRead noise corresponds to 153000 e⁻ well depth, with 16 bit ADC. ^bDark error was measured from 20 dark frames. Spatial variance was consistent across all dark frames. ^cPercent error is based on the average intensity of the plasma images in respective bands. ^dTotal error includes upper bound spectral error.

Electron Density

A contour plot of the full range of normalized electron density $n_e/n_{e,\max}$ calculated from normalized image intensity $I/I_{\max}$ and electron temperature is shown in Fig. C.8. Contours of constant electron density are displayed. From this it is observed that most of the dynamic image range corresponds to relatively low electron densities, as the emissivity scales strongly with electron temperature.

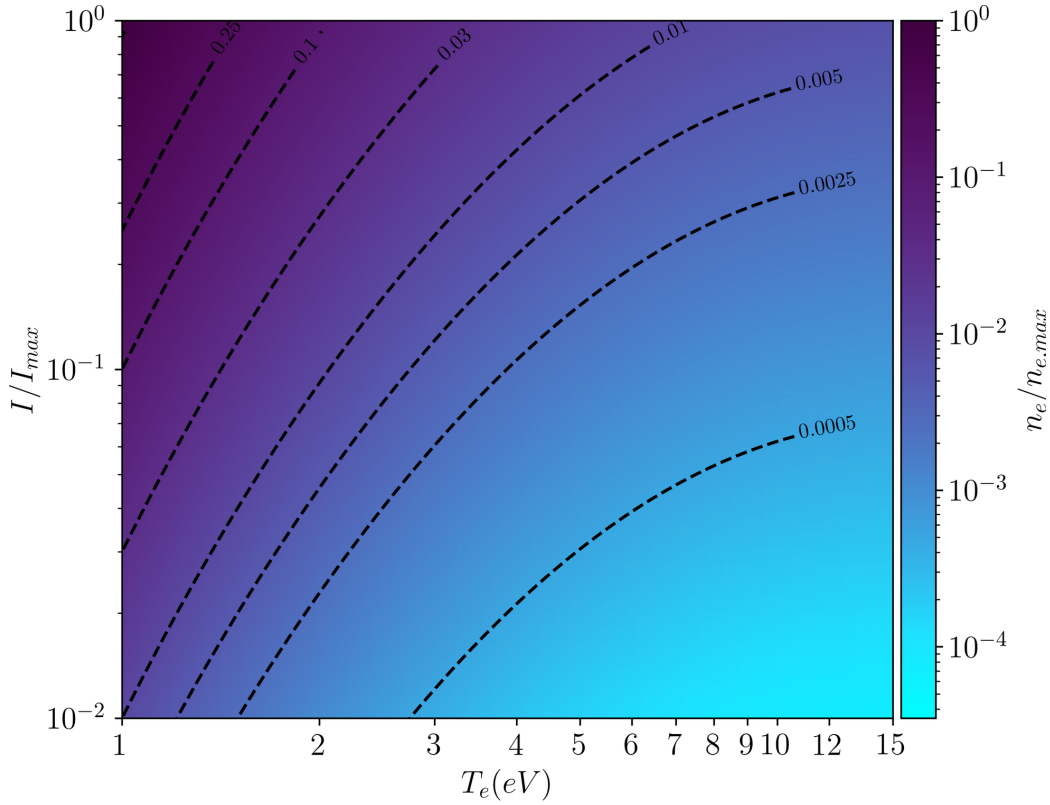


Figure C.8: Color map relating electron temperature and normalized optical intensity to reduced electron density. Contours show paths of constant reduced electron density.

Appendix D

MATHEMATICAL DERIVATIONS

D.1 Uniqueness of the Solution Vector $\vec{\xi} = (B_x, B_y, B_z, E_z)$

```
[60]: (*NV Axes*)
NV = {{1, 1, 1}, {1, -1, -1}, {-1, 1, -1}, {-1, -1, 1}} / Sqrt[3];
B = {Bx, By, Bz};
Ef = {0, 0, Ez};
(*Projections*)
Bpar = Table[(B . NV[[i]]) NV[[i]], {i, 4}];
Bperp = Table[B - Bpar[[i]], {i, 4}];
Eperp = Table[(Ef . NV[[i]]) NV[[i]], {i, 4}];

Bpar2 = Table[Bpar[[i]] . Bpar[[i]], {i, 4}];
Bperp2 = Table[Bperp[[i]] . Bperp[[i]], {i, 4}];
Eperp2 = Table[Eperp[[i]] . Eperp[[i]], {i, 4}];
Bperp4 = Bperp2^2;

system = {Gamma^2 Bpar2 + Eperp2 + Gamma^4 Bperp4 / (4 Dgs^2);
{Xi} = {Bx, By, Bz, Ez};
j = D[system, {Xi}];
```

```
[75]: detJ4atB0 = Det[j] / Ez;
Print["det(J)=", detJ4atB0 /. {Gamma -> 28025/1000000, Dgs -> 2870., Bx ->
  -418,
  By -> 782, Bz -> -2283}]
```

det(J)=4.56716

```
[77]: (*Rank, at Ez=0*)
J4atB0 = j /. Ez -> 0;
JB = J4atB0[[All, 1 ;; 3]];
Print["Rank(J)=", MatrixRank[JB]]
```

Rank(J)=3

[]:

Appendix E

SUPPLEMENTARY MATERIALS FOR WALL-INTEGRATED MAGNETOMETER

E.1 Flex PCB Antenna Design

The microwave antenna used in this experiment was based on the electrode and waveguide design initially proposed by Ma et al. (2019). From this design, the interior hole with relatively uniform microwave radiation was expanded slightly to accommodate a larger viewing angle, and the PCB was ordered from JLCPCB printed on flexible substrate.

Two versions of the PCB were used in testing. First, a basic polyimide flex PCB was used to validate the design and confirm that contrast was acceptable. This design is shown in Fig. E.1 (left). With the polyimide substrate, a significant degree of autofluorescence was observed despite the laser beam focus avoiding most of the material, decreasing contrast to about 0.7%. A version with a dyed black polyimide flex material from JLCPCB reduced autofluorescence substantially, giving our final contrast of about 1.6%. This version additionally included radial fenestrations between the concentric conductors of the antenna, as seen in Fig. E.1. These fenestrations allowed the optical adhesive to simultaneously contact each adjacent layer within the sensor assembly, and facilitated UV light into the sensor assembly for final curing of the adhesive. This version, shown on the right in Fig. E.1, was used for all presented data. Protruding wings with holes extending from the antenna allowed consistent mounting to the 3D printed chamber wall.

Electrical test points for each of the antenna rails were used to confirm proper soldering of the final connection to the SMA connector before use. Both design elements are shown in Fig. E.1. Antenna design files are available upon request.

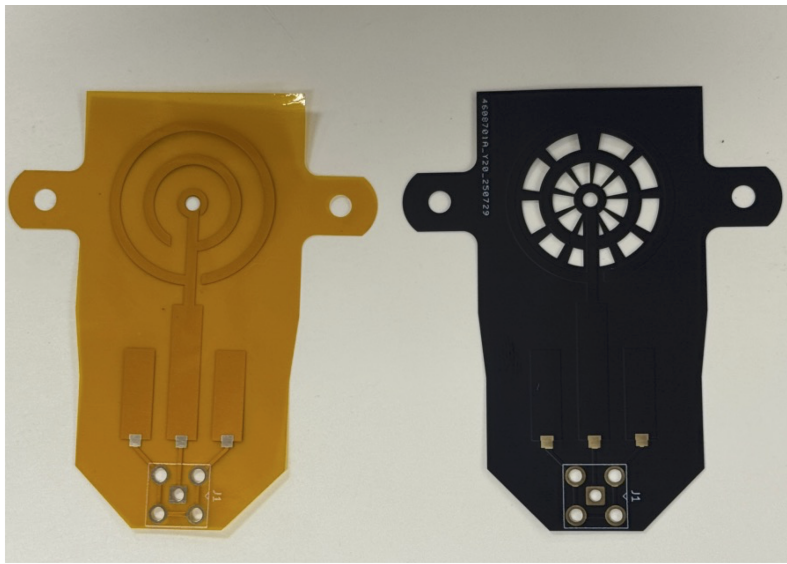


Figure E.1: Image of the polyimide flex PCB (left) and dyed and fenestrated flex PCB (right) used in the spin resonance experiments. SMA connectors not shown. Protruding wings used for repeatable mounting and conductor test points are shown.

Appendix F

ADDITIONAL METHODS

F.1 Radio Frequency Measurements

The plasma initiated on lithium niobate (LiNbO_3 , LN) in air exhibits a rich non-linear spectrum of characteristic radio frequency (RF) emission in the range of 3–100 MHz, as shown in Fig. F.1. Shear and longitudinal wave speeds (v_s, v_l) in LN are approximately 4 km s^{-1} and 7 km s^{-1} , respectively (Lunardelli et al., 2024; *Lithium Niobate* n.d.). The target dimensions are $D = 75 \text{ mm}$, $h = 0.5 \text{ mm}$, giving lowest-order mode frequencies $f_{\text{sh}} \sim 4 \text{ MHz}$ and $f_{\text{str}} \sim 7 \text{ MHz}$ for thickness-shear and thickness-stretch modes, respectively. f_{sh} appears almost exactly at the spectrum low end on LN, while the remaining peaks are generated at spacings that are integer multiples of f_{str} .

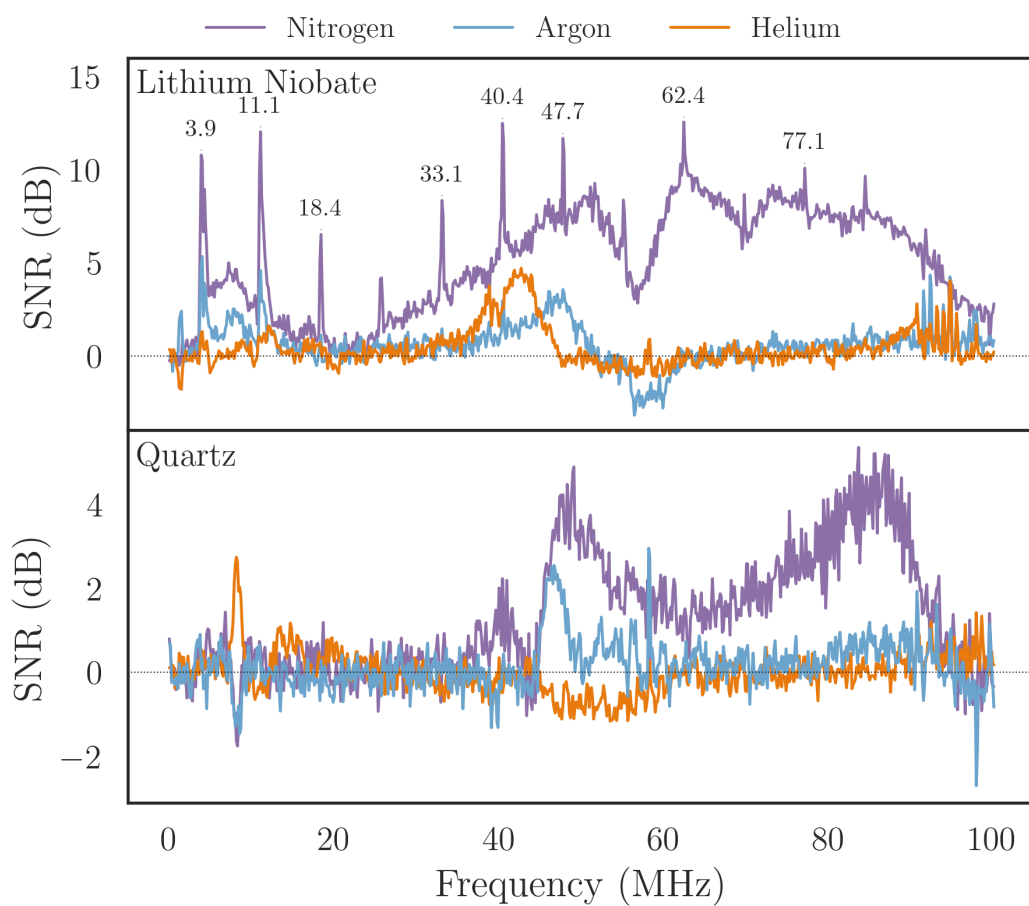


Figure F.1: Radio frequency spectra measured by coil antenna from nitrogen, helium, and argon plasma on lithium niobate (top) and quartz (bottom).