S1. Liquid Sheet Surface Imperfections

Many of the liquid sheets produced by these microfluidic nozzles (in particular the single-liquid colliding sheets but also many of the heterostructures) have noticeable surface imperfections as a result of channel roughness and the nozzle exit geometry. An example of these effects are shown in Fig. S1. These surface imperfections can be seen to not dramatically affect the interference band structure in the sheet (meaning the size of the features has to be small in comparison the wavelengths of visible light, <100 nm), but are large enough to change the local curvature of the sheet. On illumination, these structures then create bright and dark areas in the sheet’s reflection. These surface imperfections can similarly distort the ellipsometry measurements by creating reflections with different angles of incidence, which can limit the regions of the sheet which can be accurately measured.

These features do not contain noticeably more or less liquid than the rest of the sheet. This fact can be confirmed by looking at the FTIR microscopy of a single fluid sheet in comparison to the white light reflection of a sheet displaying significant surface imperfections (Fig. S1; single fluid sheet with water in all three channels: $Q_{out} = 1750 \mu$L/min and $Q_{in} = 200$...
The IR false-color image displays the intensity of the water HOH bend (1600 cm$^{-1}$) across the sheet, corresponding to the sheet thickness. The distinct surface imperfections in the white light image do not register at all in the IR microscopy data, while the IR is able to detect a thickness variation near the bottom of the sheet which is obscured in the white light image by the surface imperfections. As the height of the surface imperfections are comparable to or smaller than the noise in the IR measurements, this constrains the height of these imperfections to $\leq 35$ nm.

**Fig. S1.** White light (left) and IR false color image of a pure water sheet. The bright and dark surface imperfections in the white light image do not appear in the IR image, indicating that they correspond to only a small change in the thickness of the sheet ($\leq 35$ nm). Bottom: IR thickness data for sheet cross section indicated by dashed line.
Fig. S2. White light images of a water colliding sheet (Top) and various Water/Hydrocarbon/Water heterostructures (bottom) with toluene and cyclohexane.

While the main text focused on water sheets within hydrocarbon sheets, the heterostructures could also be generated such that a thin layer of hydrocarbon was placed within a water sheet. Stable structures were generated with both an inner toluene layer and with an inner cyclohexane layer. Sample heterostructures with $Q_{out} = 1750 \, \mu L/min$ can be seen in Fig. S2. As the interfacial tension between water and air^{2} (72 N/m) is much greater than the interfacial tension between water and toluene (36 N/m) or cyclohexane (50 N/m),^{3} at all stable flow rates examined the sheets presented the simple morphology, wherein the rims of the sheet jets overlap
and all of the liquid layers cover the same area. This is analogous to the Tol/Wat/Tol sheets with a low water flow rate presented in the main text. The water/hydrocarbon/water heterostructures still had substantially smaller surface areas than the pure water colliding sheet with the same outer fluid flow rate (Fig. S2 top), with the cyclohexane-containing sheets slightly smaller than the toluene-containing sheets. The interfacial tension between the liquids likely plays a role in the area of the heterostructures in these cases, similar to how the liquid/air interfacial tension impacts the area of the single-liquid sheets.

**S3. Stable-Unstable Transition in Heterostructures in Cy/Wat/Cy Sheets**

![Image of Cy/Wat/Cy Sheets](image)

**Fig. S3.** Unstable to stable transition for Cy/Wat/Cy heterostructures. ($Q_{out} = 1750 \mu L/min$). At low flow rates, the breakup of the small area sheet is apparent.

The morphology of the heterostructure may significantly affect the resulting emulsion. For Cy/Wat/Cy sheets, the water rims are towards the center of the cyclohexane sheet, as opposed to being coincident with the outer fluid rims in the Tol/Wat/Tol sheets shown in Fig. S3.
The water rims in the Cy/Wat/Cy sheets then disintegrate in the center of the sheet (Fig. S3), which may provide a much higher water loading in the sheet center.

S4. Additional FTIR Microscopy Images of Heterostructures

**Fig. S4.** Representative false color IR images of liquid heterostructures. Red indicates hydrocarbon and blue indicates water content. The water rims in hydrocarbon/water/hydrocarbon sheets are always within the sheet, while the hydrocarbon rims are coincident with the water rims in the water/hydrocarbon/water sheets.
Representative false color images of various heterostructure morphologies are shown in Fig. S4. Red indicates the hydrocarbon component and blue indicates the water component. In general, it can be seen that all of the hydrocarbon/water/hydrocarbon heterostructures have the water rims contained within the sheet. Even in the low-$Q_{in}$ Tol/Wat/Tol structure where the layers fill the same area, the water rims are distinct from the toluene rims. By contrast, the water/hydrocarbon/water structures have rims which overlap in the image. The exact morphology of the water/hydrocarbon/water rims seem to be more complicated, and may relate again to interfacial tension. For the water/hydrocarbon/water structures, it is energetically favorable for the hydrocarbon to form an interface with air rather than water. By contrast, in the hydrocarbon/water/hydrocarbon structures, we expect that the water energetically favors forming an interface with the hydrocarbon instead of air. However, the exact structure of the water/hydrocarbon/water rims remains ambiguous, and it is not clear that hydrocarbon/air interfaces are present at the rims of these structures.

S5. Thin Film Calculations: Transfer Matrix Method

A. Transfer Matrix Method

The transfer matrix method (TMM)\textsuperscript{4,5} was used to model the experimental data on the liquid heterostructures with IR microscopy, ellipsometry, and reflection measurements. TMM is well suited for describing the propagation of light through a stratified medium, which is the expected morphology for the liquid heterostructures. At each interface, the incident light can either transmit or reflect with coefficients given by the Fresnel equations depending on the incident polarization of light:
\begin{align*}
  r_s &= \frac{n_1 \cos \theta_1 - n_2 \cos \theta_2}{n_1 \cos \theta_1 + n_2 \cos \theta_2} \\
  r_p &= \frac{n_2 \cos \theta_1 - n_1 \cos \theta_2}{n_2 \cos \theta_1 + n_1 \cos \theta_2} \\
  t_s &= \frac{2n_1 \cos \theta_1}{n_1 \cos \theta_1 + n_2 \cos \theta_2} \\
  t_p &= \frac{2n_1 \cos \theta_1}{n_2 \cos \theta_1 + n_1 \cos \theta_2}
\end{align*} \tag{S1}

where \( n_m \) is the (complex-valued) index of refraction of the \( m \)th layer, and \( \theta_m \) is the angle of the propagation vector relative to the interface normal. \( \theta_m \) for each layer is derived from Snell’s law:

\[ n_1 \sin \theta_1 = n_2 \sin \theta_2. \] \tag{S2}

TMM uses the boundary conditions set by the interfaces with Maxwell’s equations to efficiently solve the reflected and transmitted wave amplitudes. Full derivations can be found elsewhere in the literature.\(^4\) The key result of TMM relates the forward and backward propagating wave amplitudes (\( v \) and \( w \), respectively) at the \( m \)th interface to the amplitudes at the \((m+1)\)th interface by:

\[
\begin{pmatrix}
  v_m \\
  w_m
\end{pmatrix} = M_m
\begin{pmatrix}
  v_{m+1} \\
  w_{m+1}
\end{pmatrix}
\] \tag{S3}

Where \( M_m \) is a 2x2 matrix of the form:

\[
M_m = \frac{1}{t_{m,m+1}} \begin{pmatrix}
  \exp(-i \delta_m) & 0 \\
  0 & \exp(+i \delta_m)
\end{pmatrix} \begin{pmatrix}
  1 & r_{m,m+1} \\
  r_{m,m+1} & 1
\end{pmatrix}.
\] \tag{S4}

The acquired phase difference \( \delta_m = \frac{2\pi n_m \cos \theta_m}{\lambda_{\text{vac}}} d_m \) also takes into account the layer thickness \( d_m \) and vacuum wavelength of light, \( \lambda_{\text{vac}} \). This term also accounts for absorption by the medium when the index of refraction \( n_m \) is complex-valued. The three layer, four interface liquid heterostructure reflection and transmission amplitudes can then be calculated from:
\[
\begin{pmatrix}
1 \\
r
\end{pmatrix} = \tilde{M} \begin{pmatrix}
t \\
0
\end{pmatrix}
\]
\[
\begin{pmatrix}
1 \\
r
\end{pmatrix} = \frac{1}{t_{0,1}} \begin{pmatrix}
1 \\
0
\end{pmatrix} M_M \frac{1}{t_{0,1}} \begin{pmatrix}
t \\
0
\end{pmatrix}.
\]

The matrix elements of $\tilde{M}$ then directly relate to the transmitted and reflected amplitudes of the full heterostructure, with $t = 1 / \tilde{M}_{00}$ and $r = \tilde{M}_{10} / \tilde{M}_{00}$. The transmission and reflection can be calculated for either $s$ or $p$ polarization by using the appropriate Fresnel coefficients for the reflection and transmission.

B. Application to FTIR Microscopy

As was noted in the main text, thin film interference effects produced a frequency-dependent baseline in the FTIR microscopy spectra. This effect was most prominent for Tol/Wat/Tol heterostructures, due to the weak absorption of toluene in the infrared region examined and the large area of the heterostructures compared to the Cy/Wat/Cy heterostructures. Correcting for the oscillatory baseline is also most important for structures with water as the interior liquid, as the strongest water mode (the OH bend at $3400 \text{ cm}^{-1}$) spans several hundred wavenumbers, which makes it impractical to approximate the absorption baseline as locally flat.

The FTIR measurement was performed in transmission, where the transmitted intensity through the heterostructure for a given wavelength of light is:

\[
T = |r|^2.
\]  

(For this case, the transmitted intensity’s dependence on index of refraction and angle of incidence drops out as the initial and final media are both air. As a result, there is no impedance mismatch between the final and initial medium). This intensity can be readily converted into an absorption value, $A$, by taking the logarithm:

\[
A = -\log_{10}(T).
\]
The full absorption spectrum can be fully modeled by calculating $A$ for each wavelength using the complex-valued index of refraction for each liquid as a function of frequency.\textsuperscript{6,7} As the sheets were measured at a normal angle of incidence, the $s$ and $p$ Fresnel coefficients are identical, so polarization does not need to be considered.

The oscillatory background is only apparent if the light is coherent: as the layer thickness becomes large relative to the wavelength of light, the fringes become close enough together that they blend together due to minor thickness variations, propagation angle variations, etc. As the layers get thicker, the interference background transitions from an oscillatory coherent background to a flat incoherent background. This effect was accounted for by breaking the absorption spectrum into dispersive and absorptive parts. The coherent, oscillatory background, $A_{bg}$, was calculated using the TMM method using the real-valued portion of the index of refraction. The experimental absorption spectrum was then modeled as:

$$A = A_{inc} + fA_{bg} + A_{abs}$$

Where $A_{inc}$ is the frequency-independent incoherent baseline, $A_{bg}$ is the frequency-dependent coherent baseline, $f$ is a scalar ranging from 0 to 1 to account for the coherence length, and $A_{abs}$ is the IR absorption spectrum for an incoherent film. This formulation does not account for absorption that occurs on internal reflections within the heterostructure, but the liquids used are only weakly reflecting at normal incidence. The resulting error from this simplification should be on the order of 1%.

A sample fit to an experimental spectrum is shown in Fig. S5 (Tol/Wat/Tol structure with $Q_{out} = 1750 \mu$L/min and $Q_{in} = 300 \mu$L/min, blue curve in Fig. 4A in main text). The red dashed curve is the full fit with Eq. S8, while the blue dashed curve is the baseline from $A_{inc} + fA_{bg}$. The
blue dashed curve was subtracted to get the baseline corrected absorption spectra shown in Fig. 5A in the main text.

**Fig. S5.** Accounting for the oscillatory IR background from thin film interference. Red dashed curve is the fit to the experimental spectrum with Eq. S8, while the blue curve indicates the dispersive baseline that was subtracted to generate the pure absorptive spectra in Fig. 5 of the main text.

**C. Application to Ellipsometry**

The ellipsometric angles Δ and Ψ are readily calculable using TMM. After \( \rho = r_p r_s \) are calculated for the heterostructure, the angles can be calculated from:

\[
\Psi = \arctan(\rho) \\
\Delta = -i \log \left( \frac{\rho}{|\rho|} \right).
\]  

(S9)

For the liquid heterostructures, the values of both Δ and Ψ have fine structure which arises in part from the dark bands: the points in the sheet where the four reflections fully destructively interfere. At these points, \( \rho = 0/0 \) and the ellipsometric angles are undefined. To construct the
reference curves used to describe $\Psi$ in the main text, the values of $\Psi$ at the points of maximum constructive interference were used and interpolated between. The reflectivity and full calculated $\Psi$ value for the case shown in the main text (Tol/Wat/Tol structure with $Q_{\text{out}} = 1750 \, \mu\text{L/min}$ and $Q_{\text{in}} = 300 \, \mu\text{L/min}$ at a 70° AOI) is demonstrated in Fig. S6. The reference curves shown in Fig. 8B were generated by interpolating between the values of $\Psi$ at the points of maximum reflectivity in alternating bands. (Fig. S6, red and black curves). The experimental data was similarly analyzed at the points of maximum reflectivity, as that provided the highest signal to noise.

**Fig. S6.** Calculated reflectivity and $\Psi$ values for the Tol/Wat/Tol structure. $Q_{\text{out}} = 1750 \, \mu\text{L/min}$ and $Q_{\text{in}} = 300 \, \mu\text{L/min}$ at 70° AOI. The fit curves used in the main text was generated by interpolating between the values of $\Psi$ at the maximum reflectivity for alternating bands.
D. Application to White Light Reflectivity

To generate the white light color bar shown in Fig. 9, first the reflected intensity from the sheet was calculated for each visible wavelength:

\[
I_{\text{ref}} = R I_{\text{in}} \\
I_{\text{ref}} = |r|^2 I_{\text{in}}
\]

The value of \(I_{\text{in}}\) for each wavelength was weighted based on the spectrum of the white light LED used. As was the case for the FTIR background, the white light images were taken at near-normal incidence, so the polarization effects can be neglected. Each wavelength was then converted to an RGB value using a color-matching function (CIE 1964 10-degree CMF) at a 1 nm wavelength resolution. The CMF is a function which accepts a wavelength as input and returns coefficients for red, green, and blue channels. The calculated RGB values seen for a heterostructure with given layer thicknesses could then be determined using:

\[
I_{R} = \frac{\sum_{\lambda} \text{CMF}_R(\lambda) I_{\text{ref}}(\lambda)}{\sum_{\lambda} I_{\text{ref}}(\lambda)}
\]

with corresponding expressions for the G and B channels.

An example full 2D color chart as a function of inner and outer layer thicknesses for a Wat/Tol/Wat heterostructure can be seen in Fig. S7A. In general, it can be seen that a very thin layer (< 100 nm) does not reflect any light, while a very thick layer (> 5 \(\mu\)m) reflects essentially white light, while layers around 1 \(\mu\)m thick give vibrant colors due to interference. Notably, a thick outer layer with a thin inner layer will produce a colored band against the white background corresponding to the thin inner layer. A thin inner layer is then noticeable in an otherwise thick part of a liquid heterostructure.
**Fig. S7.** Example of white light reflection off a heterostructure. (A) Characteristic reflected color of a white LED off an ideal heterostructure as a function of inner and outer layer thickness. (B) White light reflection off a Wat/Tol/Wat heterostructure with $Q_{out} = 1750 \mu$L/min and a range of $Q_{in}$. Thin film interference from a thin inner layer in a thick outer layer is visible near the top of the sheet (cf. bottom middle of B).

These characteristic colored bands of a thin inner layer could be generated in liquid heterostructures with low inner fluid flow rates. An example is shown in Fig. S7B for Wat/Tol/Wat liquid sheets. Near the top of the nozzle, the center line of the outer colliding sheet is at its thickest. In this case, the sheet is sufficiently thick that it would produce an essentially white reflection (e.g., bottom left of Fig. S7A). Introducing the inner liquid just above the threshold flow rate for generating a stable structure, the first colored band (dark yellow, bottom center of Fig. S7A) becomes visible near the top of the sheet (Fig. S7B left). As the inner flow...
rate increases, additional colored bands become visible near the top of the sheet, until at high flow rates the top of the sheet appears white again.

S6. Polarization Modulation Imaging Ellipsometry Methods

A. Working Principle

The PM imaging ellipsometer was used based on the design by Chou et al. The liquid sheet was illuminated with light polarized at 45°, meaning the incident light had equal s and p polarization components. The light reflected from the sheet is collected by an objective, after which it passes through a quarter-wave plate on a motorized stage and a linear polarizer set at 90° before reaching the CMOS camera. The field seen on each pixel of the camera can be described using Jones calculus:

\[
\begin{bmatrix}
E_{\text{ref},p} \\
E_{\text{ref},s}
\end{bmatrix} = |E_m| \begin{bmatrix}
0 & 0 \\
0 & 1
\end{bmatrix} \begin{bmatrix}
\cos^2(\beta) + i \sin^2(\beta) & (1 - i) \sin(\beta) \cos(\beta) \\
(1 - i) \sin(\beta) \cos(\beta) & \sin^2(\beta) + i \cos^2(\beta)
\end{bmatrix}
\begin{bmatrix}
|r_p| & 0 \\
0 & |r_s| e^{i\alpha}
\end{bmatrix} e^{-i\pi/4},
\]

where \( \beta \) is the angle of the quarter wave plate’s fast axis relative to the plane of incidence. The intensity seen by each pixel as a function of quarter-wave plate angle is then:

\[
I_{\text{ref}}(\beta) \propto \frac{1}{4} \left[ 3|r_s|^2 + |r_p|^2 + \left( |r_s|^2 - |r_p|^2 \right) \cos(4\beta) - 2|r_s||r_p| \left( 2\sin(\Delta) \sin(2\beta) + \cos(\Delta) \sin(4\beta) \right) \right].
\]

The ellipsometric angles \( \Delta \) and \( \tan \Psi = (|r_p|/|r_s|) \) can be determined using just five quarter-wave plate angles: \( \beta = 0^\circ, 22.5^\circ, 45^\circ, 67.5^\circ, \text{ and } 135^\circ \):
\[
I_0 = I_{\text{ref}}(0^\circ) = |r_s|^2 \\
I_1 = I_{\text{ref}}(22.5^\circ) = \frac{1}{4} \left( 3|r_s|^2 + |r_p|^2 - 2|r_s||r_p|\left(\sqrt{2}\sin\Delta + \cos\Delta\right) \right) \\
I_2 = I_{\text{ref}}(45^\circ) = \frac{1}{2} \left( |r_s|^2 + |r_p|^2 - 2|r_s||r_p|\sin\Delta \right) \\
I_3 = I_{\text{ref}}(67.5^\circ) = \frac{1}{4} \left( 3|r_s|^2 + |r_p|^2 - 2|r_s||r_p|\left(\sqrt{2}\sin\Delta - \cos\Delta\right) \right) \\
I_4 = I_{\text{ref}}(135^\circ) = \frac{1}{2} \left( |r_s|^2 + |r_p|^2 + 2|r_s||r_p|\sin\Delta \right)
\]

which can be straightforwardly algebraically manipulated to get the desired quantities:

\[
\Psi = \arctan \left( \frac{I_2 + I_4 - I_0}{I_0} \right) \\
\Delta = \arctan_2 \left( (I_4 - I_2), 2(I_1 - I_3) \right)
\]

where \( \arctan_2 \) is the two-argument arctangent function.

The results in Eqs. S14 and S15 differ from the results presented in Chou et al.\(^8\) as the linear polarizer before the camera was set to 90° instead of 0°. This means the \( r_s \) and \( r_p \) terms switch places in Eq. S13 and S14, which then changes the expression in Eq. S15. This modification was made to reduce the dynamic range seen by the camera when the sheet was near Brewster’s angle. In the Chou et al. configuration, the \( \beta = 0^\circ \) image is almost completely dark near Brewster’s angle. In the configuration used here, the sheet illumination is more uniform between the collected quarter-wave plate angles.

**B. Image Processing**

Two significant artifacts were found in the collected images. First, dark regions did not read as identically zero, likely from a diffuse scattered light background and noise on the camera. This background does not impact the calculation of \( \Delta \), as it depends only on intensity differences, but it does impact \( \Psi \). The image intensities were baselined to zero by selecting a region of the image which did not contain the sheet and subtracting the mean intensity of the region. Second,
the rotation of the quarter-wave plate was not perfectly on-axis, resulting in a translation of the image by a few pixels between different $\beta$. This translation was corrected by observing a microscope grid distortion target while rotating the quarter-wave plate. The translation caused by the wave plate rotation was measured using the grid target. The measured translation which was then used to correct for the translation on the experimental data. These methods, as well as the set angles of the polarization optics and rotation stages, were validated using a step-wafer standard of known thickness and refractive index.

For displaying the ellipsometric data maps (e.g., in Figs. 7 and 8 in the main text), the sheet was highlighted relative to the dark background by using a mask. The mask was generated by weighing the sum of all five images using a sigmoid function, with parameters chosen so that the flat part of the sheet was uniformly white and the background was uniformly dark.

**S7. Microfluidic Nozzle**

![Fig. S8. Microfluidic nozzle used in this work.](image)
The microfluidic nozzle used in this work is shown in Fig. S8 (dimensions 19 x 6 x 0.8 mm). The design is derivative of the nozzle used in the prior publication on gas accelerated sheet jets\(^9\) that is now commercially sold by Micronit, with two design differences. The radius of the outer channels \(R_{\text{out}}\) is 20 \(\mu\text{m}\) instead of 25 \(\mu\text{m}\), which results in thinner outer layers per Eq. 1. The nozzle also features etched glass filters (triangular features in Fig. S8) which can mitigate clogging of the channels. The liquids are supplied to the ports on the face of the chip and exit the nozzle at the tapered end where the etched channels converge, as shown in Fig. 1.

**S8. Secondary Sheets**

Similar to the standard colliding sheets, the liquid heterostructures generate second sheets after the cylindrical rims come together in a node at the bottom of the primary sheet. This secondary sheet is in the plane orthogonal to the primary sheet and is smaller than the primary sheet (300-500 \(\mu\text{m}\) long and about 100-200 \(\mu\text{m}\) across for the flow rates examined). The second sheet morphology was found to be extremely sensitive to \(Q_{\text{in}}\). Example structures consisting of water and toluene are shown in Fig. S9 as illuminated with the 630 nm LED lamp at a 20° angle of incidence. For small \(Q_{\text{in}}\), the second sheet was found to either be disintegrating (e.g., Tol/Wat/Tol) or otherwise poorly formed (e.g., Wat/Tol/Wat). At intermediate \(Q_{\text{in}}\), a structure closely analogous to the primary sheet liquid heterostructures were formed, where rims of (presumably) separate liquids were generated. Unlike the primary heterostructure, these sheets reflected diffusely, and the interference fringes that were visible did not clearly display patterns indicative of layered behavior. These sheets may then be emulsive (e.g., Fig. 2) as opposed to layered. At large \(Q_{\text{in}}\), the secondary sheets begin to disintegrate. For the Tol/Wat/Tol sheets, the outer sheet clearly disintegrates first, leaving an inner sheet of (presumably) water mostly intact due to its higher surface tension. Due to their smaller lengths and widths, narrow stably operating
flow rates, and less clear structure, it seems unlikely that these secondary sheets would serve as interesting spectroscopic targets. However, they may be of interest for elucidating the fluid dynamics of the entire liquid structure.

**Fig. S9.** Secondary sheets from Wat/Tol/Wat and Tol/Wat/Tol heterostructures ($Q_{\text{out}} = 1750 \mu\text{L/min}$) illuminated by a 635 nm LED at several representative flow rates.

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