Ratiometric Determination of Droplet Morphology Defined Luminescence as a Sensing Platform

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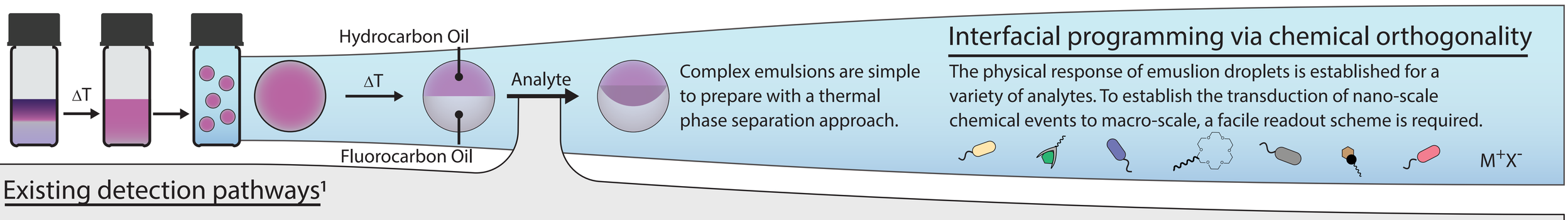
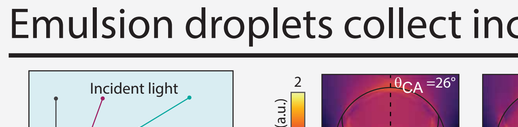
Sensors are stimuli responsive materials. Complex emulsion droplets respond to changes in chemical information as morphological transformation, characteristically.

We explore one pathway for multiplexed, reference-free, and ratiometric measurement of emulsion droplet programmable reconfiguration via concentrating and directing fluorescent light.[1,2]



Existing detection pathways[1]

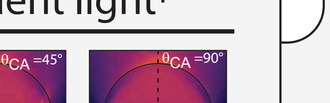
Surfactant chemistry



Pavlovic, M., et al., Anal. Chem. 2021, 93, 27, 9390-9396

Trinh, V., et al., ACS Sens. 2022, 7, 5, 1514-1523

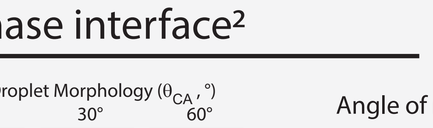
Covalent chemistry



Fong, D., et al., J. Am. Chem. Soc. 2021, 143, 11, 4397-4404

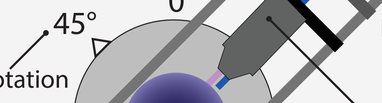
Dynamic covalent chemistry

Zeininger, L., et al., ACS Cent. Sci., 2019, 5, 5, 789-795



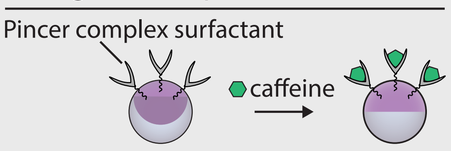
Zentner, C. A., et al., J. Am. Chem. Soc., 2019, 141, 45, 18048-18055

Enzyme or acid cleavable



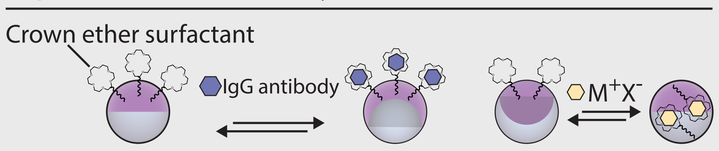
Zarzar, L. D., et al., Nature 2015, 518, 520-524
Zarzar, L. D., et al., PNAS 2017, 114, 15, 3821-3825

Host-guest complexation



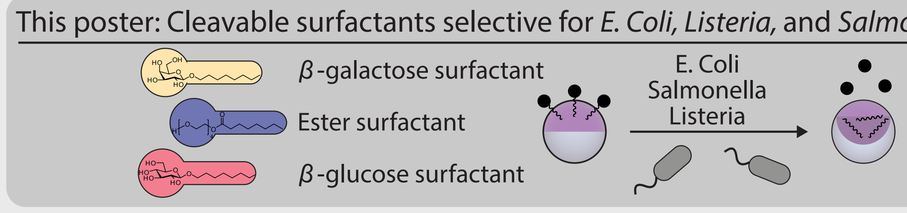
Zeininger, L., et al., Lab Chip 2019, 19, 1327-1331

Supramolecular chemistry



Djalali, S., et al., Adv. Funct. 2021, 32, 2, 2107688

This poster: Cleavable surfactants selective for *E. Coli, Listeria,* and *Salmonella*[3]

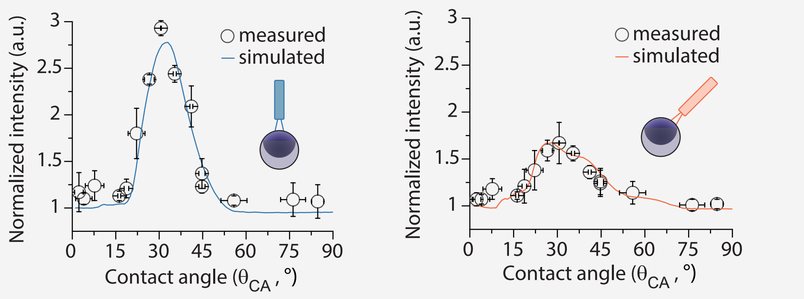


Emulsion droplets collect incident light[1]

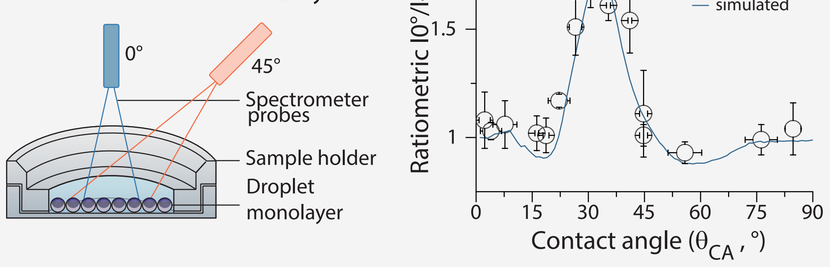
Light totally internally reflects along the internal hydrocarbon-fluorocarbon interface, dependant on refractive indices and droplet morphology.

Droplet monolayers: a reference-free sensor[2,3]

We monitor the fluorescence of layers of dyed Janus emulsion droplets.

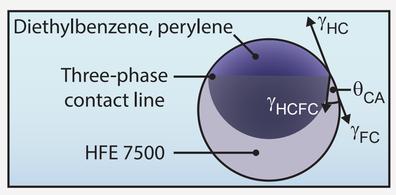


By monitoring droplet emission at two angles, fluorescence is recorded ratiometrically.

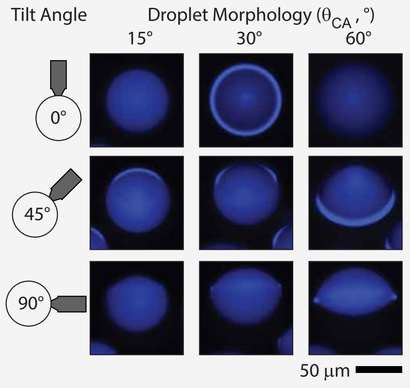


Ratiometric morphology determination is analyte chemistry-independent, and enables polydisperse emulsions for on-site application: translating nano-scale chemical events into macro-scale fluorescence signal.

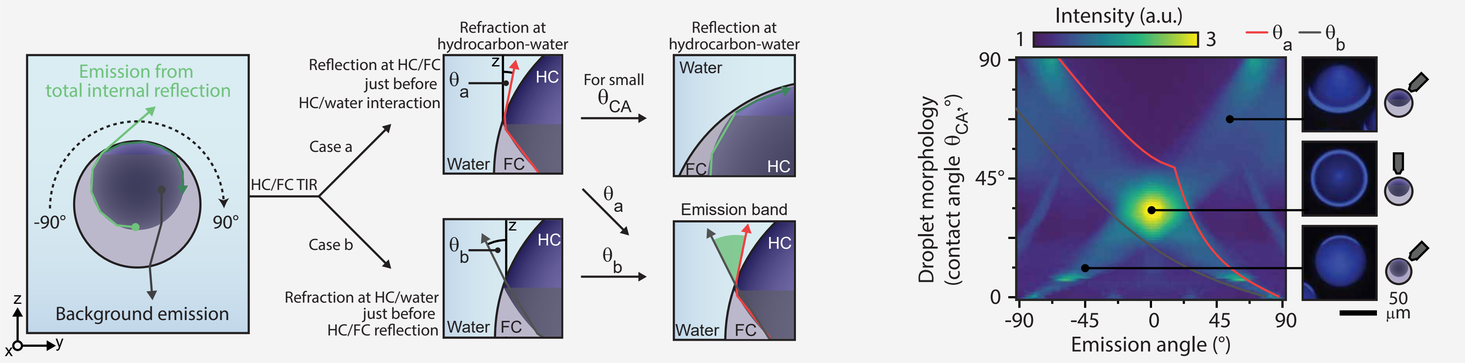
Light is directed by the three-phase interface[2]



When a fluroescent dye is placed within the hydrocarbon phase, an annular fluorescent ring can be viewed only at particular angles.

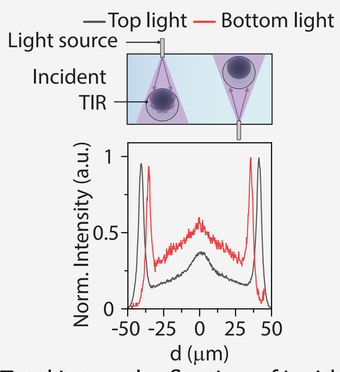


Complex droplets align with gravity, a rotating fluorescence microscope is used to image droplets at any angle.

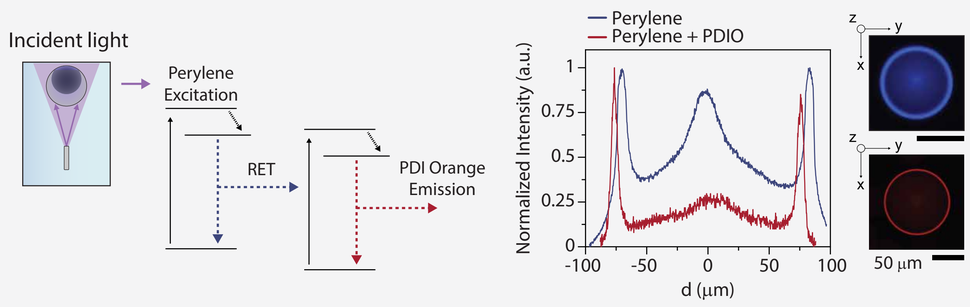


Angular emission can be calculated analytically, and is modelled via ray tracing, displaying a verifyable trend.

Collaborative optical effects enable further design



Total internal reflection of incident light enhances fluorescence concentration.

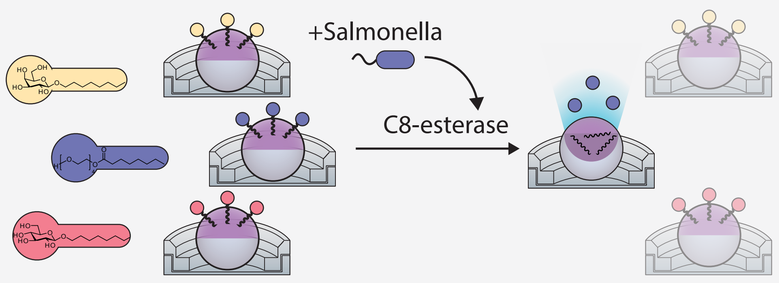


We include a second dye in the hydrocarbon phase to collect the concentrated perylene emission, enhancing the signal-to-noise ratio of the emission band.

Droplets as foodborne pathogen sensors[3]

E. Coli → β-galactosidase
Salmonella → C8-esterase
Listeria → β-glucosidase

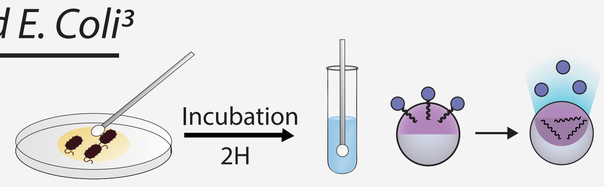
We target three major foodborne bacteria which release characteristic enzymes. Cleavable surfactants act as targeted enzymatic substrates.

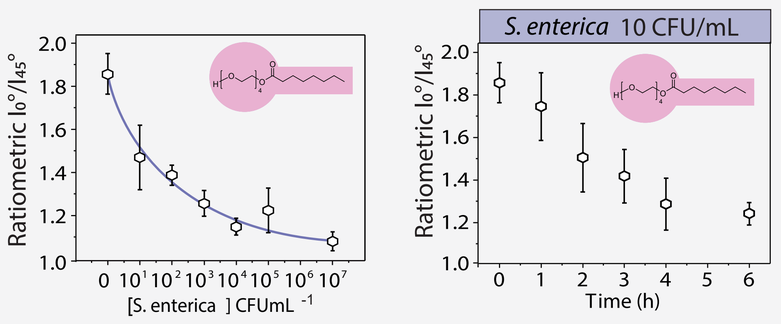


Ratiometric detection of droplet fluorescence serves as a chemistry-independent readout for polydisperse emulsion samples.

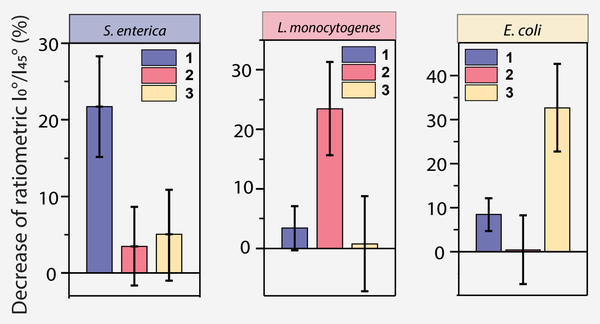
Ratiometric detection of *Salmonella, Listeria, and E. Coli*[3]

1. Bacteria are incubated in substrate solutions.
2. After a period of time, droplets are prepared in the solution
3. Ratiometric fluorescence intensity is tested





C8-esterase released by Salmonella cleaves the substrate, demonstrating sensitivity to 10 bacterial cells per mL after 2 hours.



Cross tests and swab tests demonstrate high sensitivity and specificity toward targeted pathogens, and with ON-OFF behavior for deployment.

1. Pablo Simón-Marqués, Bradley D. Frank, Aleksandr Savateev, Lukas Zeininger, Adv. Opt. Mater 2021
2. Bradley D. Frank, Sara Nagelberg, Agata W. Baryzewska, Pablo Simón-Marqués, Markus Antonietti, Mathias Kolle, and Lukas Zeininger, Submitted
3. Agata W. Baryzewska, Christian Roth, Peter H. Seeberger, Markus Antonietti, and Lukas Zeininger, Submitted

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