Electronic Supplementary Information for the Manuscript "Infrared imaging with visible light in microfluidic devices: the water absorption barrier" by Mona Suryana,<sup>a</sup> Thomas Produit,<sup>b</sup> Hongzhi Yang,<sup>b</sup> Giovanni Birarda,<sup>c</sup> Jegan Vishnuwardhana Shanmugar,<sup>a</sup> Leonid Krivitsky, <sup>b</sup> Anna Paterova,<sup>b,†</sup> and Gianluca Grenci \*a,d

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#### 1. Microfluidic device fabrication protocol

#### Step 1: Preparation of the SU-8 master mold

#### 1.1 Pattern definition

Please refer to Table S1 for the processing parameters of each mold.

- 1.1.1 Dehydrate the silicon wafer at 200°C for 15 min.
- 1.1.2 Spin coat SU-8 photoresist on the silicon wafer.
- 1.1.3 Soft bake the photoresist on a hot plate.
- 1.1.4 Expose the photoresist to UV light (with i-line filter, 365nm) through the first photomask.
- Apply post exposure bake to the exposed photoresist on a hot plate. 1.1.5
- 1.1.6 Develop the photoresist using a SU-8 developer at room temperature, rinse with isopropyl alcohol, and blow dry with nitrogen.

#### 1.2 Silanization of the SU-8 molds

- 1.2.1 Treat the molds with oxygen plasma at 60 W for 30 s with 20 sccm of oxygen flow and 1-10 mbar chamber pressure.
- 1.2.2 Place the molds in a vacuum jar with approximately 50µL of Trichloro(1H,1H,2H,2Hperfluorooctyl) silane and leave the jar in vacuum (1-10 mbar) for at least 2 hours.

Process step	Master mold #1	Master mold #2	Master mold #3
	Thickness 15.6 µm	Thickness 28 µm	Thickness 47 μm
Spin coat	SU-8 3010:	SU-8 3025:	SU-8 3050:
	1200 rpm for 30 s	2600 rpm for 30 s	2600 rpm for 30 s
Soft bake	95°C for 10 min	95°C for 15 min	95°C for 20 min
UV exposure dose	150 mJ/cm <sup>2</sup>	150 mJ/cm <sup>2</sup>	150 mJ/cm <sup>2</sup>
Post exposure bake	65°C for 1 min, 95°C	65°C for 1 min, 95°C	65°C for 1 min, 95°C
	for 3 min	for 4 min	for 3 min

Table S1: Processing parameters for SU-8 master molds.

#### Step 2: Preparation of PDMS secondary mold

- 1.1 Thoroughly mix Sylgard 184 PDMS elastomer and curing agent with 10:1 ratio.
- 1.2 Degas the mixture for approximately 15 min or until there are no visible bubbles.
- 1.3 Pour the mixture on the SU-8 master mold prepared in Step 1 and degas the PDMS mixture.
- 1.4 Cure the PDMS on a hot plate at 65°C for 1.5 hours.
- 1.5 Peel off the PDMS replica from the SU-8 mold.
- 1.6 Silanize the PDMS replica with the same steps as the SU-8 master mold in Step 1.

## Step 3: Fabrication of the second half of the microfluidic device

3.1 Substrate preparation

- 3.1.1 Treat a silicon wafer with oxygen plasma (60 W, 30 s, 20 sccm of oxygen flow, 1-10 mbar chamber pressure).
- 3.1.2 In an e-beam evaporator, deposit 10 nm chromium adhesion layer, followed by 50 nm gold layer.
- 3.1.3 Spin coat Hexamethyldisilazane (HMDS) adhesion layer on the silicon wafer at 3000 rpm for 30 seconds.
- 3.1.4 Spin coat AZ 5214E photoresist at 3000 rpm for 40 seconds, followed by soft bake at 100°C for 1.5 min.
- 3.1.5 Expose the photoresist to UV light through the second photomask with energy dose of 140 mJ/cm<sup>2</sup>.
- 3.1.6 Develop the photoresist in AZ 400K developer diluted with 1:4 ratio, rinse with deionized water, and gently blow dry with nitrogen. The measured pattern thickness is approximately 1.5 μm.
- 3.1.7 Remove the HMDS layer by oxygen plasma treatment for 20 seconds.
- 3.1.8 Etch the gold layer using gold etchant diluted with 1:1 ratio for 40 seconds, rinse with deionized water, and blow dry with nitrogen.
- 3.1.9 Etch the chromium layer using chrome etchant for 20 seconds, rinse with deionized water, and blow dry with nitrogen.
- 3.1.10 Remove the AZ 5214 photoresist by soaking in acetone and IPA, followed by oxygen plasma for 2 min.

## 3.2 SU-8 pattern preparation

- 3.2.1 On the gold-patterned substrate prepared earlier, spin coat SU-8 3010 photoresist at 1400 rpm for 30 seconds, followed by soft bake at 95°C for 10 min.
- 3.2.2 Expose the SU-8 to UV light (with i-line filter, 365nm) through the third photomask with energy dose of 150 mJ/cm<sup>2</sup>.
- 3.2.3 Apply post exposure bake to the SU-8 on a hot plate at 65°C for 1 min, followed by 95°C for 3 min.
- 3.2.4 Develop the SU-8 using a SU-8 developer, rinse with isopropyl alcohol, and blow dry with nitrogen. The measured pattern thickness is approximately 14.5  $\mu$ m, i.e. close to, but not higher than the height of the thinnest microfluidic chamber.
- 3.2.5 Hard bake the SU-8 at 120°C for 2 min.

## 3.3 Dicing of the completed silicon wafer

Cut the silicon wafer to the size of the final device: 40x25mm.

# Step 4: Fabrication of the first half of the microfluidic device C1

- 4.1 Prepare a PDMS wall by cutting out a PDMS sheet (1 mm thick, same as the CaF<sub>2</sub> thickness) in the center area with the size of the device and the inlet and outlet reservoirs for NOA73 dispensing.
- 4.2 Prepare a Kapton film with a cut-out circular hole in the center (11 mm diameter, slightly bigger than  $CaF_2$  window, which has 10 mm diameter).
- 4.3 Place the PDMS secondary mold (prepared in step 2) on a flat surface, e.g. a glass plate, with the pattern facing up.
- 4.4 Place the PDMS wall on the PDMS mold, aligned with the border of the microfluidic device layout.
- 4.5 Treat the CaF<sub>2</sub> window with oxygen plasma (60 W, 30 s, 20 sccm of oxygen flow) to improve NOA73 flow during the following fabrication.

- 4.6 Place the CaF<sub>2</sub> window on the PDMS mold, with the microfluidic chamber aligned with the center of the window.
- 4.7 Place a PDMS sheet (1 mm thick) on top of the PDMS wall and CaF<sub>2</sub> window to cover the center cavity, while ensuring that the size of the PDMS sheet does not block the inlet and outlet reservoirs for the NOA73 dispensing.
- 4.8 Dispense NOA73 gradually at the inlet reservoir and let it fill the cavity. When NOA73 reaches the window edge, capillary flow will fill the thin gap between the PDMS mold and the window.
- 4.9 After the device cavity is completely filled, place the Kapton film prepared earlier on top of the flat PDMS, with the circular hole aligned with the  $CaF_2$  window.
- 4.10 Expose the assembly to UV light (UV LED exposure system, 365nm) to partially cure the NOA underneath the window. With power density of 38 mW/cm<sup>2</sup> and 80% power, it requires 1 min of exposure.
- 4.11 Carefully remove the Kapton film and leave the assembly alone for 15 min.
- 4.12 Expose the assembly to UV light again to finish the NOA curing. With the same settings, it requires 1.5 min of exposure.
- 4.13 Remove the flat PDMS and PDMS wall from the assembly, then carefully remove the cured NOA layer from the PDMS mold.
- 4.14 Punch through a hole (1.5 mm diameter in our case) at each the inlet/outlet feature in the NOA layer to create inlet and outlet connections for the device.

Note: There is some shrinkage of NOA73 upon curing, causing slightly lower NOA73 pattern thickness (15  $\mu$ m) compared to the SU-8 master mold (15.6  $\mu$ m).

## Step 5: Fabrication of the first half of the microfluidic device C2 and C3

- 5.1 Prepare PDMS by thoroughly mixing Sylgard 184 PDMS elastomer and curing agent with 10:1 ratio, and degasing the mixture for approximately 15 min or until there are no visible bubbles.
- 5.2 Repeat steps 4.1 to 4.8, except that Kapton sheet is not used (skip step 4.2) and NOA73 is replaced with the prepared PDMS in step 5.1. Note that the flat PDMS and PDMS wall must be silanized in this case while it is not necessary if NOA73 is used.
- 5.3 After the device cavity is completely filled, cure the PDMS at room temperature for 1 day, followed by 65°C for 1.5 hours on a hot plate. Curing first at room temperature avoids air bubbles which may form at elevated temperature.
- 5.4 Remove the flat PDMS and PDMS wall from the assembly, then carefully remove the cured PDMS layer from the PDMS mold.
- 5.5 Punch through a hole (1.5 mm diameter in our case) at each the inlet/outlet feature in the PDMS layer to create inlet and outlet connections for the device.

Note: Similarly, there is some shrinkage of PDMS upon curing, causing slightly lower PDMS pattern thickness (26 and 44  $\mu$ m) compared to the SU-8 master mold (28 and 47  $\mu$ m). The first half of devices C2 and C3 can also be fabricated using NOA73, just like C1.

## Step 6: Bonding of the two halves of the device

- 6.1 Treat the first and second halves of the microfluidic devices with oxygen plasma (60 W, 30 s, 20 sccm of oxygen flow).
- 6.2 Bring both halves of the device facing each other, such that the corresponding features in both halves are aligned.
- 6.3 Gently press both halves at the corner to fix the position of the two halves.
- 6.4 Cut an 8 mm diameter PDMS cylinder (1 mm thick) and place it centered on top of the  $CaF_2$  window.

- 6.5 Place a PTFE sheet (0.5 mm thick, the size of the device) on each side of the assembly.
- 6.6 Place the assembly in a mechanical press.
- 6.7 For device C1, apply 150kg load, gradually heat from room temperature to 100°C, and hold for 30 min. After it has cooled down to room temperature, remove the device.
- 6.8 For devices C2 and C3, apply 25kg load and remove the device after 1 day.

Note: For devices made using NOA73, bonding should be done on the same day, preferably as soon as the first half of the device (i.e. NOA layer with embedded  $CaF_2$  window) is completed. Late bonding would result in poor bonding strength between the two halves.



**Figure S1:** Side view schematics of the fabrication protocol for the first half of microfluidic device C1.



2. Step height measurement of the microfluidic layer before assembly, using stylus profiler

**Figure S2:** Step height measurement of microfluidic layer C1 (NOA73), C2 (PDMS), and C3 (PDMS) fabricated in step 4 and 5, before assembly. Dashed blue arrow: scan direction.



**Figure S3:** Reflectance spectrums of the devices for confirmation of the thickness of the chambers C1 (15 $\mu$ m), C2 (26 $\mu$ m) and C3 (44 $\mu$ m), after assembly. The estimated chamber thicknesses are shown on the right of the graphs.

## 3. Optical measurement of the microfluidic chamber after assembly

The optical measurements are performed in Reflectance mode, with the layer stack of the materials set as follows:

Category	Material	Thickness
		(μm)
Misc	Air	Infinite
Dielectrics	CaF <sub>2</sub>	1000
Misc	Air	To be measured
Metals	Au	0.05
Metals	Cr	0.01
Semiconductors	Si	Infinite

**Table S2:** Layer stack of the materials in the device during reflectance measurement.

The air in the middle of the stack refers to the chamber thickness (for the tool to estimate), while the air at the top layer of the stack refers to the environment above the sample and is taken as having

infinite thickness in the calculation by the tool. Likewise, the substrate (in this case Silicon) is also taken as having infinite thickness. The refractive index of each layer material is recorded in the materials library in the software as Cauchy parameters or raw data, i.e. refractive index vs. wavelength.

As seen from Table S3, the thicknesses measured using stylus profiler and optical reflectometer are comparable.

Device	Thickness (µm)		
	Stylus profiler, before assembly Optical reflectometer, after assembl		
C1	14.7	14.6	
C2	26	27.4	
C3	43.9	46.2	

**Table S3:** Comparison of the approximate thickness of the microfluidic layer/chamber before/after assembly.

### 4. Explanation of the spectra extraction from Q-IR (Figure 4 in the text)

Since the magnification in quantum imaging depends on the signal and idler wavelength ratio  $\lambda_{signal} / \lambda_{idler}$  [1], one must rescale each raw Q-IR data to intercompare images across different idler wavelengths  $\lambda_{idler}$ . Hence, each image frame in Figures 4 and 5 is obtained by rescaling the raw Q-IR images by a zoom factor ( $\lambda_{idler} - \lambda_{Pump}$ )/ ( $\lambda_{idler,0} - \lambda_{pump}$ ), where  $\lambda_{idler,0} = 2815$  nm is taken as the arbitrary reference scale and  $\lambda_{pump} = 532$  nm is the pump laser wavelength.

The absorption spectrum shown in Figure 4(e) in the main text, it is obtained by the following procedure:

- From the visibility of the interference is directly proportional to the complex amplitude transmission coefficient of the sample  $V_{x,y} = (I_{max} I_{min})/(I_{max} + I_{min})^{\sim} |\tau_{IR}|$ . Thus, by taking the ratio of the visibilities of interference pattern at the region with sample and without sample (reference), we can find out the transmissivity of the sample  $|\tau_{IR}|$ . However, since setup utilizes a Michelson interferometry scheme, the probe photons pass through the sample twice. As a result, our measurements directly yielded the intensity transmissivity T (denoting intensity transmission within a single pass through the sample) [2].
- From the rescaled images for each of the five colored regions (black, red, blue, green, pink) highlighted in the image frames above the SU-8 patterns, we average the ratio of visibility values  $V_{x,y}$  from the region of interest to the reference value  $V_{white,\lambda}$ . This procedure is repeated for every idler wavelength to reconstruct a spectrum for each colored region:

$$T_{color, \lambda} = \frac{1}{N} \sum_{(x,y) \in colored region} \frac{V_{x,y}}{V_{white, \lambda}},$$
  
where  $T_{color, \lambda}$  is the computed transmission for the colored region  
region at idler wavelength  $\lambda,$   
 $V_{white, \lambda} = \frac{1}{N} \sum_{(x,y) \in white region} V_{x,y},$  and  $N$  is the total number of pixels in the colored/white regions.

- The absorption  $A_{color, \lambda}$  at a particular idler wavelength  $\lambda$  is then simply given by:

$$A_{color, \lambda} = 1 - T_{color, \lambda}.$$

- The final absorption spectra  $A_{color final}$ , given as the collection of  $A_{color, \lambda}$  for each  $\lambda$ , is obtained via renormalizing each  $A_{color, \lambda}$  by dividing to the maximal value:

$$A_{color\,final, \lambda} = A_{color, \lambda} / \max_{\lambda} A_{color, \lambda}$$



### 5. Visibility plots through 44 $\mu$ m water layer

**Figure S4:** (Left) Visibility map of the interference pattern obtained for microfluidic device C3 at 3390 nm probe wavelength. (Right) Vertical cross sections for visibility maps (average of data in between orange lines) for water layer in device C3 (44  $\mu$ m) for different wavelengths. From the displayed curves, the significant reflection through water starts from **3390 nm**.



#### 6. FTIR spectra of SU-8 in water-filled C1, C2 and C3 devices

**Figure S5:** Raw data of the FPA imaged integrated at 3017nm, the red square represents the areas used in Figure 5. A single FPA tile is 170x170 microns, C1 and C2 maps are a mosaic of 4x4 tiles, C3 is a mosaic of 5x5 tiles. Scale bars are 200 microns and colour bar is from 0 to 2.0 a.u.



**Figure S6:** Absorption spectra of SU-8 structures acquired by mean of FTIR in the C1, C2 and C3 devices when filled with water.

7. Extraction of absorption spectra from Q-IR of microfluidic devices filled with water



**Figure S7:** Example of a transmittance image obtained after rescaling of visibility Q-IR images. The white region shows the chosen pixel which sees only absorption from water. The pixels in the red region see both SU-8 and water absorption.

For QIR images the visibility of the interference is determined by the amplitude transmission of the idler photons through the sample. Since we use Michelson interferometry system, the photons propagate twice though the medium layers. Thus, the visibility is directly proportional to the intensity transmission of the idler photons. In this case, for each idler wavelength  $\lambda$  the transmission T(x,y) though microfluidics chip is given by the Beer-Lambert law:

$$T_{red}(x,y,\lambda) = e^{-L_{water\ above\ SU8}(x,y)\alpha_{water\ }(\lambda)} T_{SU8}(x,y,\lambda) \quad (1)$$
$$T_{white}(x,y,\lambda) = e^{-L_{water}(x,y)\alpha_{water\ }(\lambda)}, \quad (2)$$

where  $T_{red}(x,y,\lambda)$  is the region with SU8 and water layers in microfluidic chip,  $T_{white}(x,y,\lambda)$  is the region with only water layer,  $L_i(x,y)$  is the depth at pixel (x,y) and  $\alpha_i(\lambda)$  is the absorption coefficient at the wavelength  $\lambda$ ,  $T_{SU8}(x,y,\lambda)$  is the transmission through SU-8 below the water layer. First, we obtain the wavelength dependent absorption coefficient for water layer  $\alpha_{water}(\lambda)$  from Equation (2) and averaging over the region of interest, shown by white square:

$$\alpha_{water}(\lambda) = \frac{1}{N} \sum_{(x,y) \in white \ region} - \frac{\log[T_{white}(x,y,\lambda)]}{L_{water}}$$

where *N* is the number of pixels in the region of interest, and  $L_{water} = 15 \,\mu m$  for C1,  $L_{water} = 26 \,\mu m$  for C2 and  $L_{water} = 44 \,\mu m$  for C3.

Next, from Equation (1) we can find out the transmission thought the SU-8 layer as:

$$T_{SU8}(x,y,\lambda) = \frac{T_{red}(x,y,\lambda)}{T_{white}(x,y,\lambda)}e^{-\alpha_{water}(\lambda)(L_{water}(x,y) - L_{water above SU8}(x,y))},$$

where and  $L_{SU8} = 14.5 \,\mu m$ ,  $L_{water above SU8}$  is 0.5  $\mu m$ , 11.5 and 29.5  $\mu m$  for C1, C2 and C3, respectively.



Figure S8: SU-8 absorption spectra for microfluidic devices C1, C2 and C3

Finally, we calculate the absorption spectra as  $A(x,y,\lambda) = 1 - T(x,y,\lambda)$ , shown in Figure S8



### 8. Vertical cross sections of the IR images

**Figure S9:** Plots for vertical cross sections in FTIR measurements over SU-8 pattern in devices C1, C2 and C3 for **3429 nm** and **3314 nm** wavelengths. Dashed lines correspond to signal levels over SU-8 pattern with water layer on top (red) and water layer only (black).

**Figure S10:** Plots for vertical cross sections (for *x* pixels from 85 to 100 in the camera) in Q-IR microscopy over SU-8 pattern in devices C1, C2 and C3 for **3429 nm** and **3314 nm** wavelengths. Dashed lines correspond to signal levels over SU-8 pattern with water layer on top (red) and water layer only (black). From these cross sections we obtain ~4  $\mu$ m spatial resolution of the method, where we defined the resolution as the 10-90% rise/fall from maximum measured visibility.



#### 9. Material table

Chemical	Company	Product no.	Comments
Trichloro(1H,1H,2H,2H- perfluorooctyl)silane 97%	Sigma Aldrich	448931-10G	
Hexamethyldisilazane	Sigma Aldrich	379212	
Gold etchant	Sigma Aldrich	651818	Diluted with deionized water to 1:1 ratio
Chrome etchant	Sigma Aldrich	651826	

Sylgard 184 Silicone Elastomer Kit	Dow Corning		Polydimethylsiloxane or in short, PDMS
Norland Optical Adhesive 73	Norland Products Inc.	7304	
SU-8 3010 photoresist	MicroChem	Y311060	
SU-8 3025 photoresist	MicroChem	Y311072	
SU-8 3050 photoresist	MicroChem	Y311075	
SU-8 developer	MicroChem	Y020100	
AZ 5214E photoresist	Merck	14744719710	
AZ 400K developer	Merck	21433126644	
Acetone	Fisher Scientific	A949-4	
Isopropanol	Fisher Scientific	P/7507/17	
Material	Company	Product no.	Comments
Silicon wafer, 4 inch, prime grade	Bonda Technology Pte Ltd		
CaF2 IR-grade windows	Crystran, UK	CAFP10-1	10 mm diameter, 1 mm thickness
Kapton film (DuPont)	RS Components Pte Ltd	536-3952	
Equipment	Company	Product no.	Comments
UV-KUB 2 (UV LED exposure system)	KLOE		Emission spectrum 365nm ± 5nm
CEE Spin coater	Drawar Calanaa		
	Brewer Science	Model 200x	
NJB4 mask aligner	SUSS MicroTec	Model 200x	
Precision digital hot plate	SUSS MicroTec Harry Gestigkeit GmbH	Model 200x 2860SR	
Precision digital hot plate Plasma Surface Technology	SUSS MicroTec Harry Gestigkeit GmbH Diener Electronic GmbH + Co. KG	Model 200x 2860SR	For O <sub>2</sub> plasma treatment
Precision digital hot plate Plasma Surface Technology Electron beam evaporator	SUSS MicroTec Harry Gestigkeit GmbH Diener Electronic GmbH + Co. KG AJA International Inc	Model 200x 2860SR	For O <sub>2</sub> plasma treatment
MJB4 mask alignerPrecision digital hot platePlasma Surface TechnologyElectron beam evaporatorBruker IFS 66v/s	SUSS MicroTec Harry Gestigkeit GmbH Diener Electronic GmbH + Co. KG AJA International Inc Bruker	Model 200x 2860SR	For O <sub>2</sub> plasma treatment

Table S4: List of chemical, material, and equipment used in the device fabrication and characterization.

## References

[1] G. B. Lemos *et al.*, "Quantum imaging and metrology with undetected photons: tutorial", *J. Opt. Soc. Am. B* **39**, 2200 (2022).

[2] A. V. Paterova *et al.,* "Tunable optical coherence tomography in the infrared range using visible photons", *Quantum Science and Technology* **3**(2), 025008 (2018).