VOLTAGE PROGRAMMABLE LIQUID OPTICAL INTERFACE

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1. Device Configuration – Solid Dielectric

In the work presented in the manuscript two different solid dielectric layers were used directly on top of the electrodes.

The first was a dielectric stack layer consisting of 17 alternating thin film layers of silicon dioxide and titanium dioxide with a total thickness of 1.13 µm. It was designed to specification and deposited by Yorkshire Photonic Technology Limited, Halifax, U.K. The dielectric stack was designed to be transparent at a wavelength of 633 nm and to be strongly reflecting at 543 nm. The wrinkle amplitude was visualised and measured at 633 nm in transmission in the interferometer whilst simultaneously observing the diffraction of light in reflection at 543 nm due to the periodicity of the wrinkle. A device with this type of dielectric stack was used for the measurements shown in figures 2 and 3 of the manuscript. For figure 3 this enabled simultaneous observation of the time dependent diffraction pattern and measurement of the amplitude of the surface wrinkle at the times between the step voltage changes. This allowed us to ensure that a constant wrinkle amplitude of $A = 0.36$ µm was used for the dynamic measurements at the 3 different oil film thicknesses.

The second was a photoresist layer fabricated from a commercial material, SU-8 (SU8 formulated in GBL, chemically amplified epoxy based negative resist. MicroChem Corporation, 1254 Chestnut Street, Newton, MA 02464, USA). Devices with this type of dielectric layer were used for the transmission measurements shown in figures 4 and 5. The processing of this layer has been described in detail in reference [1]. The main steps in the process are: (i) spin coat SU8-10 in a 50% solution of its own developer (spreading stage followed by 2000 r.p.m. for 30 seconds); (ii) soft bake at 65ºC for 1 minute and 95 ºC for 3 minutes; (iii) flood

expose to UV at 50 mW cm^{-2} for 6 seconds; (iv) postbake at 65 ºC for 1 minute and 95 ºC for 1 minute; and (v) harden at 175 ºC for 30 minutes.

2. Liquid surface waves created by vibration

The use of diffraction or scattering of light from liquid surface waves created by vibration has been used historically to measure the liquid surface tension of the liquid [2] and more recently to investigate the damping of low frequency surface acoustic waves [3,4]. In comparison to our approach, where the fluid in a wrinkle is static, the fluid is in constant motion and the diffraction pattern, recorded at near grazing incidence depends on the distance from the "exciter" that is used to vibrate the surface of the fluid.

3. Properties of Oils

There are a range of other physical properties that can be selected to optimise device performance; a selection of possible oils and their properties are listed in table 1. To maximise the amplitude of a wrinkle for a given applied voltage requires a high dielectric constant and low surface tension. However, for a sinusoidal phase profile the intensity of a transmitted zero order is reduced to zero when the difference between the maximum (peak) and the minimum (trough) phase is given by $1.53\pi = 4.80$ radians (from the Fraunhofer approximation for a thin sinusoidal phase grating [5]). To achieve this at a lower wrinkle amplitude, and therefore also at a lower voltage, requires high refractive index oils. In addition, for fast switching response to step changes in the applied voltage a low viscosity is required. There is a compromise in selecting an oil that has the optimum combination of physical properties, is colourless (at the wavelength used), does not evaporate too quickly, and is non-toxic. From table 1, 1-decanol is a good compromise in this respect. If the commercial RI matching oils are polar and have dielectric constants that are bigger than the square of the refractive index (due to Langevin type contributions) then they are likely to be promising, but at restricted wavelengths because they have some coloration.

4. Extending to Two-Fluid Systems

Our devices involve producing a surface wrinkle on the oil-air interface of a single layer of oil. Such liquid based devices are restricted in their orientation of operation

by gravity. However, it is possible that the surface wrinkle could be produced at the interface between two density matched liquids, e.g. oil-water, in an encapsulated device [6]. This type of approach is used in the Varioptic variable focus liquid lens to produce devices which can be used in any orientation and which are robust to shock and mechanical vibrations [7,8]. The disadvantage of this approach is that there is a smaller difference between the refractive indices of oil and water relative to the large difference between the refractive indices of oil and air. Therefore to produce the maximum diffraction efficiency (e.g. to reach the minimum in the transmission of the zero order undeviated beam) the wrinkle amplitude at an oil-water interface would need to be several times larger than the amplitude that would be required at the oil-air interface. Many oils have quite different refractive index to water, but are very similar in density (Table 1).

5. Scaling Results for Equation 1

Equation 1 provides a scaling relationship for the amplitude of a periodic wrinkle to the voltage, V_o, and the film thickness-to-pitch ratio ($\bar{h}/p$) in the form,

$$A = \left(\frac{\alpha(\varepsilon_{oil} - \varepsilon_{air})}{\gamma} \right) V_O^2 \exp\left(-\frac{\beta\bar{h}}{p} \right) \qquad (S1)$$

This equation was derived for an applied voltage on a film of oil directly on top of the electrodes by considering the change in the dielectric energy stored in the liquid and the interfacial energy stored in an assumed sinusoidal wrinkle of the liquid-air interface above the electrodes and minimising with respect to the wrinkle amplitude. The assumptions in deriving eq. (S1) are,

(i) the wrinkle amplitude is small ($A << p$)

(ii) the periodic potential profile due to the electrodes, $V(x, y)$, is described by a Fourier series expansion to second order only

(iii) the potential profile is unperturbed by the presence of the oil/air interface

(iv) the oil-air interface for a wrinkle is sinusoidal.

For a given electrode width and gap, a Fourier series expansion of the voltage profile can be made and the dielectric energy calculated [9]. To obtain an analytically

tractable result this was used in the calculation of the dielectric energy $W_E = \frac{1}{2}\mathbf{D}\cdot\mathbf{E}$, terms to second order were retained, and the energy was then integrated over the fluid volume above the electrodes. The integral involves a series expansion for the case where the wrinkle amplitude is much less than the electrode and gap widths. The net result is an energy that depends on V_o^2, multiplying terms involving a factor with the wrinkle amplitude, and exponentials containing wrinkle amplitude to pitch (we used equal mark-space ratio). The wrinkling of the liquid-air interface changes its area and this can be calculated for small amplitudes. The combination of these two energies was then minimized with respect to changes in wrinkle amplitude.

6. Validity of Equation S1

Equation S1 provides good design insight into the various physical properties needed to achieve the photonic effects, but experimentally we found a need for a calibration factor in the exponential. The theory has a number of approximations (as described above) to keep it analytically tractable and produce a formula with the key dependencies correctly described (in terms of confirmation by experiment). The number of Fourier modes is truncated and the complexities introduced by the dielectric layer and the distortion in electric field at the upper liquid-air boundary are not accounted for. In addition, the experimental configuration for the devices reported includes a thin dielectric layer between the plane containing the electrodes and the liquid; a complete theoretical treatment would require corrections to the theory described above. However, we have extensive data on decanol testing the scaling laws and further theory and the data, once we have a range of liquids, will form the basis of a subsequent report. For decanol, the experiments have included multiple electrode pitches (40, 80, 120 and 180 μm) and oil film thicknesses between 9.8 μm and 25.1 μm. We have so far found the following experimental scaling relationship holds for a range of values of the oil film height h and pitch, $p=2d$:

$$\log_e\left(\frac{A}{V_o^2}\right) = 6.34 + 2.705\left(\frac{\bar{h}}{d}\right) \tag{S2}$$

The analysis does appear to correctly identify dependences on particular parameters, for example obtaining the ratio of the dielectric constant to the surface tension in the coefficient of proportionality gives intuitive insight into how to increase the wrinkle amplitude for a given voltage. Equation S1 provides design insight into the various

physical properties needed to achieve the photonic effects; the need for a calibration factor in the exponential is not a fundamental problem in the photonics concept.

References

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Table 1: Physical Properties of a Selection of Candidate Oils

Oil	Dielectric Constant	Viscosity (Ns m^{-2})	Surface Tension (mN m^{-1})	Refractive Index	Density (g cm^{-3})
Hexadecane[a]	2.05	0.00303	27.47	1.4345	0.773
1-decanol[a]	8.1	0.01130	28.4	1.438	0.830
Glycerol[a]	47.2	0.945	63.4	1.473	1.26
Nitrobenzene*[a]	34.8	0.00178	43.9	1.55	1.2
PDMS 47V20	2.68[b]	0.019[c]	20.6[c]	1.400[b]	0.950[c]
PDMS 47V350	2.80[b]	0.340[c]	21.1[c]	1.403[b]	0.970[c]
Immersion oil[d]	N/A	0.10-0.12	N/A	1.515	1.025
RI Matching fluid*[e]	N/A	0.015	29	1.460-1.570	0.832
RI matching fluid*[e]	N/A	0.085	37	1.570-1.640	1.006

N/A : data is not available for this parameter

(a) Knovel Critical tables, 2nd ed. online (2009).

(b) Bluestar Silicones France SAS, Rhodorsil Oils 47 (http://www.bluestarsilicones.com).

(c) Fanton, X., Cazabat, A.M. & Quéré, D. *Langmuir* **12**, 5875-5880 (1996).

(d) Merck Cat. No. 1.04699 "Immersion oils for microscopy".

(e) Cargille labs, RI matching liquids series A (http://www.cargile.com/refractivestandards.shml).

　　*Coloured liquid.