Supplementary material (ESI) for Journal of Materials Chemistry

SUPPLEMENTARY INFORMATION

Formation of 3-D optical and structural lattices with 2 mutually incoherent white light beams in a photopolymerisable material

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Materials and Methods

Sample preparation

As detailed elsewhere[1], photopolymerisable organosiloxane was prepared through acid-catalyzed hydrolysis and condensation of 3-(trimethoxysilyl) propyl methacrylate (MAPTMS, Gelest, Inc., PA, USA). In a typical procedure, 1.1 g (5.5×10^{-5} mol) of 0.05 N hydrochloric acid (HCl, diluted from standardized 0.1 N HCl aqueous solution from Sigma-Aldrich, Canada) was added to 17.6 g (0.0708 mol) of MAPTMS in a glass phial. The initially phase-separated mixture homogenized in 5 minutes to a transparent sol with negligible absorption in the visible region (400 to 800 nm). The sol was sensitized to visible light with 0.05 wt. % of the free-radical photoinitiator (bis(η^5cyclopentandienyl) bis(2,6-difluoro-3-(1H-pyrrol-yl)-phenyl) titanium(IV) (λ_{max} = 393 nm, 460 nm, Ciba Specialty Chemicals Inc., Canada). The mixture was shielded from ambient light, stirred for 20 hours and filtered through a polytetrafluoroethylene (PTFE) membrane (0.2 μm pore size, Pall Corporation, USA) prior to use.

Sample characterisation

After lattice formation, solid organosiloxane samples were wholly extracted from glass cuvettes. The pale yellow, transparent cubes were cleaved along the [100], [010] or [001] faces. Samples were examined under an optical microscope (Carl Zeiss Inc.) in reflection mode or sputter-coated with a thin Au layer and imaged through a scanning electron microscope (Phillips 515).

[1] Burgess, I. B., Shimmell, W. E. and Saravanamuttu, K. Spontaneous Pattern Formation Due to Modulation Instability of Incoherent White Light in a Photopolymerizable Medium. *J. Am. Chem. Soc.*, **129**, 4738-4746 (2007).

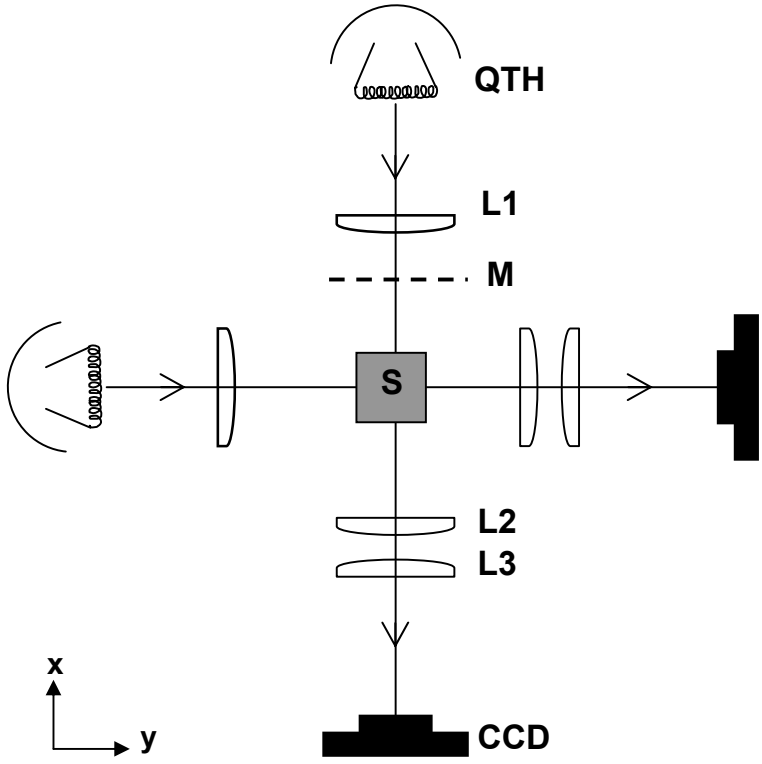

Figure S1 Schematic of optical assembly for 2-beam configurations The assembly consisted of two orthogonal beams of white light emitted by two separate quartz-tungsten-halogen (QTH) lamps. Each beam was collimated by a plano-convex lens (L1) and launched into a transparent cuvette containing the polymerisable organosiloxane (S). The spatial intensity profile of each beam at the exit face of the sample was imaged by a pair of plano-convex lenses (L2,L3) onto a CCD camera. An amplitude mask (M) was placed in the path of one or both beams.

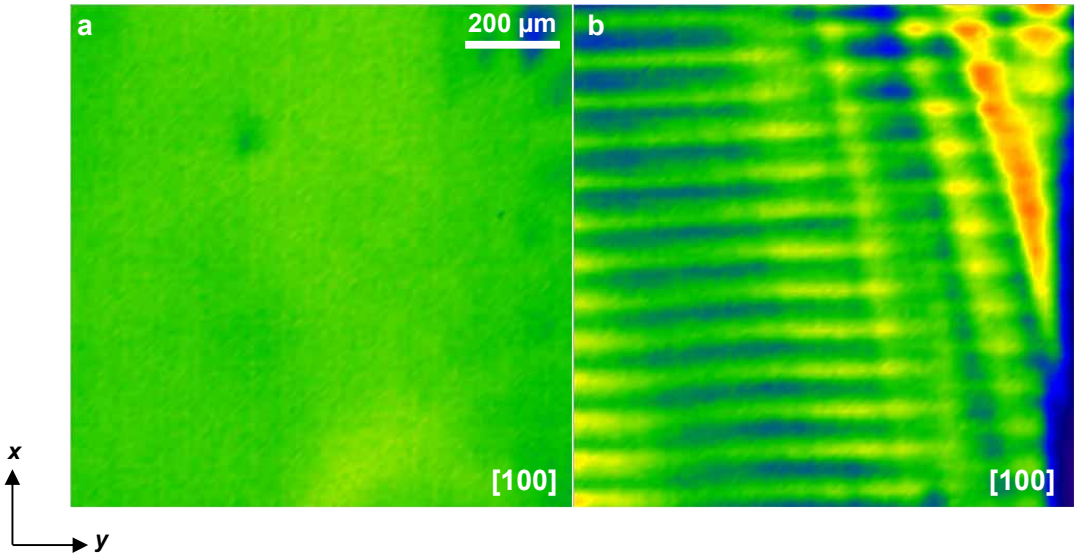

Figure S2 Spatial intensity profiles of a passive beam X (> 550 nm) in configuration X+Yz. The initially uniform profile of beam X (a) develops a periodic modulation of bright stripes (b), confirming that it divides and propagates as self-trapped lamellae within the *same* periodic stack of lamellar waveguides induced by beam Yz. The same scale applies to both images.

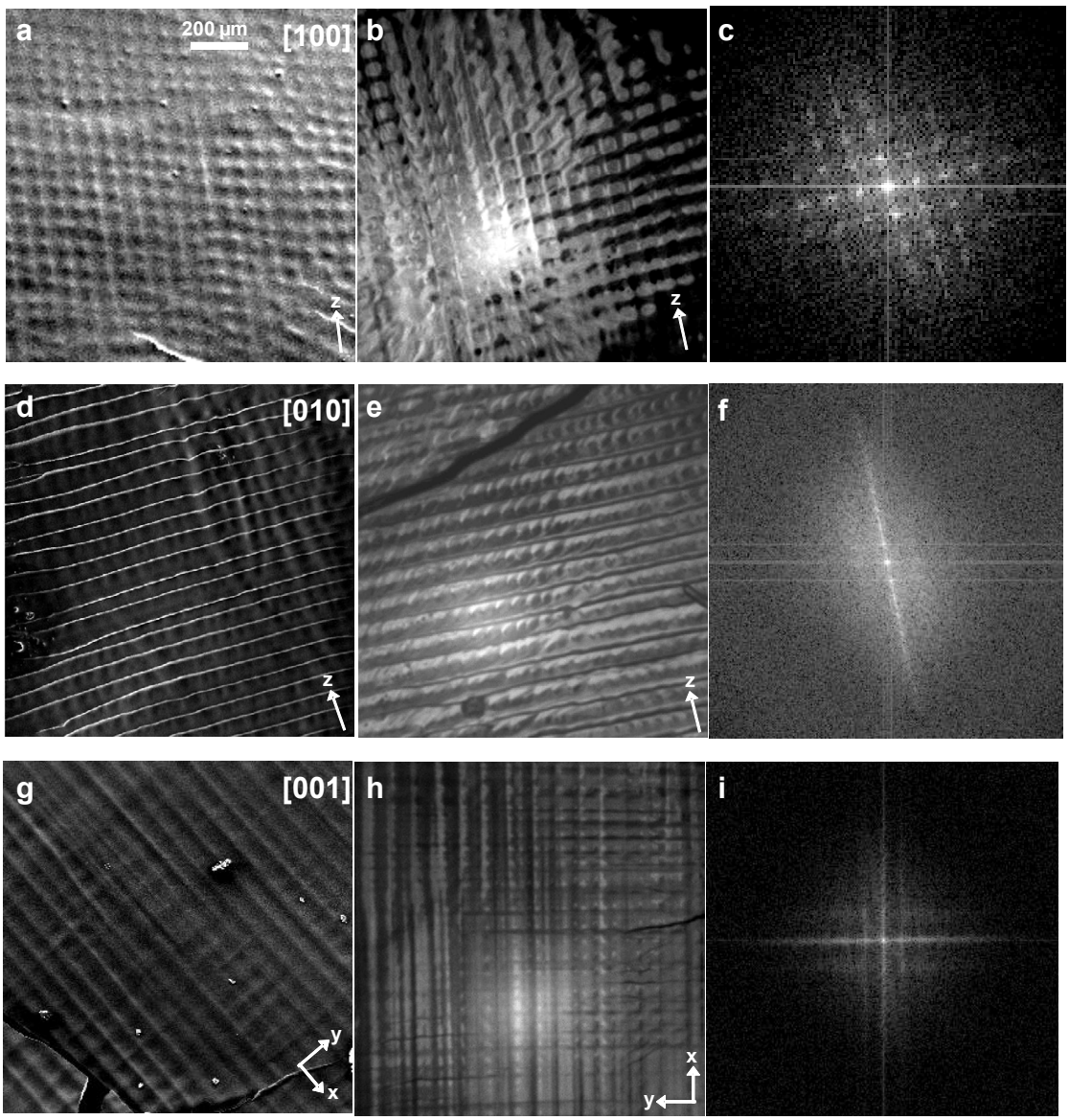

Figure S3 Lattice structure induced by 2-beam configuration X_y+Y_z. Scanning electron and optical micrographs and Fourier transforms of the optical micrographs are shown for the [100] (a-c) [010] (d-f) and [001] (g-i) lattice faces. The scale bar in (a) applies to all micrographs.

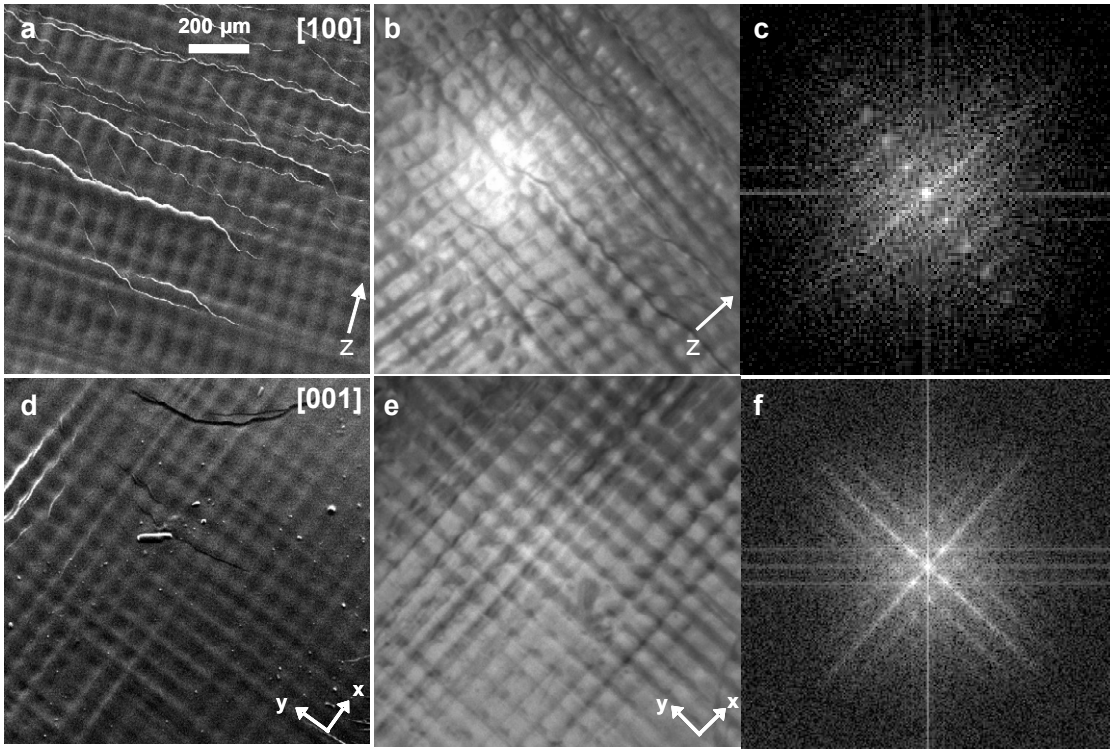

Figure S4 Lattice structure induced by 2 -beam configuration X_y+Y_x Scanning electron and optical micrographs and Fourier transform of the optical mic rograph are shown for the (a-c) [100] and (d-f) [001] lattice faces. The scale bar in (a) applies to all micro graphs.

2-beam configuration	Observation plane	Axis	σ_a	σ_P
$\mathbf{X + Y_z}$	(100)	y	0.26	0.24
		z	0.08	0.11
	(010)	x	0.26	0.27
		z	0.07	0.10
$\mathbf{X_y + Y_z}$	(100)	y	0.12	0.06
		z	0.09	0.06
	(010)	x	0.27	0.21
		z	0.09	0.08
$\mathbf{X_y + Y_x}$	{100}	y/x	0.12	0.06
		z	0.10	0.25
$\mathbf{X^*}$	(100)	z	0.28	0.11
		y	0.27	0.12
$\mathbf{X_z}$	(100)	z	0.11	0.04
		y	0.26	0.36

Table S1 Analysis of the regularity of the 3-D and 2-D lattices of self-trapped filaments induced by 2 beam and single beam configurations, respectively. For each configuration, the normalised standard deviation of the alignment (σ_a) and periodicity (σ_P) is given for the two transverse axes of each observation plane. The values were obtained from averages from at least thrice-repeated experiments. *In the case of the 2-D lattice by beam X alone, periodic lines were arbitrarily imposed on the array during analysis giving artificially excellent values of periodicity. The poor values of alignment however show that the self-trapped filaments were randomly positioned along both transverse axes.