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15	Organic light-emitting transistors are pivotal components for emerging opto- and
16	nano-electronics applications, such as logic circuitries and smart displays. Within
17	this technology sector, the integration of multiple functionalities in a single
18	electronic device remains the key challenge. Here we show optically switchable
19	organic light-emitting transistors fabricated through a judicious combination of
20	light-emitting semiconductors and photochromic molecules. The irradiation of the
20	solution-processed films at selected wavelengths enables the efficient and
22	reversible tuning of charge transport and electroluminescence simultaneously,
22	with a high degree of modulation (on/off ratios up to 500) in the three primary
23 24	colours. Different emitting patterns can be written and erased, through a non-
24 25	invasive and mask-free process, on a length scale of few microns in a single device,
25 26	thereby rendering this technology potentially promising for optically gated highly-
27	integrated full-colour displays and active optical memory.
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29	Organic light-emitting transistors (OLETs), combining in a single device the
30	functions of light generation of organic light-emitting diodes (OLEDs) with the current
31	modulation (and signal amplification) of organic thin-film transistors (OTFTs), have
32	emerged as a promising new class of devices with significant potential for integrated
33	optoelectronics, smart display technology, and organic lasers ¹⁻⁵ . The emitting layer of
34	the unencapsulated OLETs is easily accessible for comprehensive optical and electrical
35	investigation of the fundamental physical processes, thereby providing powerful

36 insights into device physics^{6,7}. The fabrication of OLETs does not require multiple

metal evaporation steps that are potentially damaging for the interface between 37 electrodes and active layers. As a result, the simplified device architecture enables its 38 potential use in active matrix displays. Furthermore, the position of the recombination 39 region in the channel of ambipolar OLETs can be shifted away from the electrodes as a 40 function of the applied bias, avoiding the metal-induced quenching of excitons⁸⁻¹⁰. 41 42 Capelli *et al.* recently demonstrated that the performance of OLET can be boosted by exploiting a trilayer heterostructure, surpassing the equivalent OLED efficiency over 43 100 times¹¹. OLETs with performance comparable to display pixels driven by 44 polycrystalline-silicon backplane transistors have been reported, which can be operated 45 at low-voltage and low-power consumption¹². 46

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48 Besides the efforts devoted to enhance the charge carrier mobility, efficiency, and brightness of OLETs¹³⁻¹⁶, the integration of further functionalities into a single device 49 represents another important challenge with the prospect of realizing controllable 50 integrated circuitry^{17,18}. Electrically switchable chiral light-emitting transistors (LETs) 51 have been demonstrated, in which the current direction can be used to control the 52 polarization of light from *pn* junctions in tungsten diselenide (WSe₂), which serves as 53 a channel material of LETs¹⁹. Another example is an all-graphene based light-emitting 54 field-effect device featuring an external electrical bias tuning of the emission 55 spectrum²⁰. However, these multifunctional LETs, based on two-dimensional (2D) 56 materials differ in terms of the nature of the charge transport and mechanism of light 57 generation compared to OLETs. The current approach to control "multifunctional" 58

LETs still relies on external electrical driving. Conversely, optical control offers various
advantages, such as non-invasively, high spatial and temporal resolution, and the
possibility to tune both wavelength and intensity of the emitted light²¹.

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Some recent reports demonstrated that it is possible to fabricate optically 63 switchable multifunctional OTFTs²²⁻²⁴, by blending organic semiconductors with 64 photochromic diarylethenes (DAEs). Upon exposure to light of different wavelength, 65 DAEs can be toggled between two isomers with different electronic properties, and they 66 also show high thermal stability and fatigue resistance during continuous 67 photoswitching²⁵⁻²⁷. Such optically responsive OTFTs exhibited high current switching 68 ratios and large charge carrier mobilities, and have been applied for the fabrication of 69 flexible non-volatile optical memory with over 256 distinct levels²⁸. 70

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Here we report the fabrication and characterisation of the optically switchable 72 organic light-emitting transistors (OSOLETs), by integrating DAEs into the light-73 emitting semiconducting layer of OLETs via simple solution processing. Both output 74 current and electroluminescence (EL) are simultaneously modulated by irradiating the 75 devices at distinct wavelengths. We demonstrate three classes of OSOLETs emitting 76 over the entire visible spectrum (green, red, and blue), which can reversibly and 77 remotely switch output current and EL on and off via visible and UV light irradiation. 78 In addition, emitting patterns within one pixel of the OSOLET can be written and erased 79 easily by using a light beam as an external, non-invasive, and mask-free writing tool 80

81 with a spatial resolution down to the far-field diffraction limit, i.e. $\lambda/2NA < 1 \mu m$ (with 82 λ being the wavelength of visible light and NA the numerical aperture of the optical 83 tool used). In view of current minimum pixel sizes, e.g. in the best "retina" displays 84 (~55.5 µm), the present system holds particular potential of reversibly encoding high-85 density visual information into a single pixel of a high-resolution display.

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87 Mechanism and device fabrication

Three commercially-available semiconducting light-emitting polymers poly(9,9-88 dioctylfluorene-alt-bithiophene) (F8T2), poly[2-methoxy-5-(3',7'-dimethyloctyloxy)-89 para-phenylenevinylene] (MDMO-PPV), and poly(9,9-dioctylfluorene) (F8) were 90 used to fabricate OLETs having fluorescence emission ranging from blue to red (Fig. 91 1a), thus covering the entire visible spectrum (supplementary Fig. S1 for the absorption 92 93 and photoluminescence spectra). To enable the optical switching in OLETs, an energetic matching between the photochromic molecules and the emissive materials is required, 94 i.e., the highest occupied molecular orbital (HOMO) levels of the emissive polymers 95 96 should energetically be positioned in between those of the open and closed DAEs (Fig. 1b). According to cyclic voltammetry (CV) measurements of the light-emitting 97 polymers (see supplementary Fig. S2) and previous studies on DAEs^{23,29,30}, the HOMO 98 99 levels of DAEs in their open forms are slightly below the one of the green and blueemitting hosts (~100 meV) and comfortably below that of MDMO-PPV (~200 meV). 100 Conversely, the HOMO levels of the DAEs in their closed forms are > 600 meV higher 101 than the HOMO of the light-emitting polymers in all three binary components, and thus 102 significant hole trapping is expected for the DAEs in their closed form. In view of the 103

different HOMO levels of the three polymers responsible for red, green, and blue light
emission, we have selected two DAEs, i.e., DAE_tBu and DAE_F (Fig. 1a)^{29,30}, having
high fatigue resistance over repetitive photoswitching cycles in the solid state (Fig. S4).
Based on the energetic considerations above, DAE_tBu molecules act as switchable
charge traps within the matrix of the emissive F8T2 and MDMO-PPV, whereas the
lower HOMO level of DAE_F makes it suitable in combination with the blue emitter
(F8).

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The OSOLETs were fabricated in a bottom-gate bottom-contact configuration with 112 SiO₂/Si as substrates and the pre-patterned gold interdigitated electrodes as source and 113 drain contacts (Fig. 1c, see device fabrication in the supplementary information). To 114 enhance the charge carrier mobility, self-assembled monolayers (SAMs) of octadecyl-115 trichlorosilane (OTS) were chemisorbed onto the SiO₂/Si substrates^{31,32} prior to spin-116 coating the solutions of the light-emitting polymers, followed by their thermal 117 annealing at 170 °C to leverage their optoelectronic properties³³. To avoid thermal 118 119 degradation of the DAEs at such high temperature, the photochromic molecules were dissolved in solvents orthogonal to those used for the polymers and spun on top of the 120 emissive layer. A gentle post-annealing process was applied to activate the thermal 121 122 diffusion of DAE molecules into the polymer matrix (80 °C for 1 h in the case of F8T2 and MDMO-PPV, while 40 °C for 1.5 h with regards to F8). The morphologies of the 123 deposited light-emitting polymer/DAE bicomponent films were investigated by atomic 124 force microscopy (AFM, see supplementary Fig. S3). The deposition of DAE tBu via 125 this permeation process did not modify the morphology of F8T2 and no phase 126

separation was observed. Only minor morphology variations were monitored in the case of MDMO-PPV. However, the exposure of neat F8 to both the solvent and thermal treatment, helps the diffusion of DAE_F into F8, and further promotes the formation of supramolecular structures (such as the crystalline phase³⁴ and the conformational isomer β-phase³⁵) of F8, resulting in a rougher morphology (from root-mean-squared roughness R_{rms} = 3.0 nm for the neat F8 film to R_{rms} = 12.3 nm for the F8/DAE_F film) due to the presence of micron-sized islands from the aggregation³⁶.

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We assessed the retained photoisomerization ability of DAEs in the solid state in 135 the presence of the selected light-emitting polymers for the OSOLETs. UV/visible 136 absorption spectroscopy on the light-emitting polymer/DAE bicomponent films 137 revealed upon UV (312 nm) irradiation the appearance of the typical spectral features 138 in the visible region for the closed DAE isomer that disappear upon green light (> 520 139 nm) irradiation (see supplementary Fig. S5). These observations provide unambiguous 140 evidence that after diffusion into the three polymer matrices the DAEs are still able to 141 undergo reversible photoisomerization. 142

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144 Characteristics of OSOLETs

145 The performance of OSOLETs in the three primary colours was characterised by 146 using EL spectroscopy as well as transfer curves of current density and luminance *vs.* 147 gate voltage, as summarized in Fig. 2. The light generated within the channel was 148 observed when OSOLETs were in operation, and thereby EL spectra and optical images

(inset) of the green, red and blue OSOLETs (channel length $L = 2.5 \mu m$ and channel 149 width W = 1 cm) were recorded (Figs. 2a-c). The emission peaks of the green and red 150 OSOLETs are located at 540 nm (FWHM = 90 nm) and 630 nm (FWHM = 143 nm), 151 respectively. Two emissive bands appear in the F8/DAE F based OSOLET with the 152 main peak at 450 nm. The red-shift of EL spectra, compared to that of glassy F8 film 153 with the S_1 to S_0 vibronic peaks at 420 nm, suggests a high fraction of β -phase chain 154 conformations in the F8/DAE F binary component film³⁵. Furthermore, we note the 155 presence of an emission band at longer wavelengths, i.e. a green band, which is 156 common in F8-based emissive devices due to the formation of inter-chain states and/or 157 fluorenone defects³⁷⁻⁴⁰. Importantly, the emissive bands of the three OSOLETs cover 158 well both the visible region (400-700 nm) and even stretch into the near infrared (NIR) 159 160 up to 800 nm.

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Although light emission requires bipolar injection, most of the transport of our 162 OSOLETs can be described as essentially unipolar, as inferred from the dependence of 163 the drain current on the applied gate voltage, in line with previous literature on non-164 switchable OLETs⁴¹⁻⁴³. While the holes are evenly distributed within the device channel, 165 light emission provides indeed evidence for electron injection and diffusion ranging to 166 several nm inside the channel. Yet electrons remain minority carriers up to the highest 167 (gate and drain) voltages tested here, as their contribution to the overall current is never 168 appreciable. For this reason, electron transport and light modulation thereof cannot be 169 determined by using the chosen materials (lowest unoccupied molecular orbital 170

171 (LUMO) levels listed in Scheme S1). The corresponding light is emitted closely to the 172 electrode in F8T2 based OLET with a longer channel width ($L = 20 \mu m$) (see 173 supplementary Fig. S6). However, owing to the resolution of our camera and the narrow 174 channel width of the OSOLETs, the light emission was observed over the entire channel 175 area in the optical images (Figs. 2a-c).

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The transfer characteristics of green, red, and blue OSOLETs using DAEs as photo-178 switchable units were measured (Figs. 2d-f). When the DAEs are in their open forms, 179 180 the hole mobilities extracted from the transfer characteristics in the saturation regime (see supplementary information for device characterisation) are about 1.5×10^{-3} , 1×10^{-3} 181 ⁴, and 5×10^{-7} cm² V⁻¹s⁻¹, respectively. For the sake of comparison, the field-effect 182 mobilities in pristine green, red, and blue polymers based OLETs have been measured 183 to be ca. 5×10^{-3} , 2.0×10^{-4} , and 8×10^{-5} cm² V⁻¹s⁻¹, respectively (see supplementary Fig. 184 S7). Clearly the hole mobility of F8T2 and MDMO-PPV is slightly reduced due to the 185 scattering (and/or residual trapping) of DAEs in the polymeric matrix. However, the 186 mobility in the blue OSOLETs with the open isomer of DAE F drops by two orders of 187 magnitude with respect to the one using the neat polymer. It appears that domain 188 boundaries and disordered interphase regions in the F8/DAE F bicomponent film result 189 in a much rougher morphology, thereby hindering charge transport⁴⁴. 190

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192 The luminance of the OSOLETs was collected simultaneously during measurement193 of the transfer characteristics (Figs. 2g-i), and it shows a similar trend as drain current

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194	vs. gate voltage (V_g). It should be noted that the pristine F8T2 and MDMO-PPV based
195	OLETs exhibit slightly higher EL intensity as bicomponent ones measured under
196	identical conditions (see supplementary Fig. S7). Interestingly, in the case of F8/DAE_F,
197	despite the reduction of the output current with respect to the pristine material, we
198	observe an increase of luminance, which can be ascribed to the higher
199	photoluminescence efficiency (η_{PL}) of F8 in the so-called β -phase as already reported
200	by Perevedentsev and Hsu et al ^{35,45} . Therefore, F8T2 and MDMO-PPV based OLETs
201	exhibit a minor variation of EQE, between pristine and bicomponent devices (EQE at
202	$V_g = -80 \text{ V}$ for: MDMO-PPV 2×10 ⁻³ %, MDMO_PPV/DAE_tBu 6×10 ⁻³ %, F8T2
203	0.4×10^{-2} %, and F8T2/DAE_tBu 1.0×10^{-2} %). However, the big change in the
204	morphology leads to a significant increase of the EQE (again at $V_g = -80$ V) of the
205	F8/DAE_F bicomponent device (1.2 %) when compared to the neat F8 device (1.2 $\times 10^{-10}$
206	² %). Although the controlled formation of the β -phase of F8 <i>via</i> dipping in solvent/non-
207	solvent mixtures has already been exploited to improve the EQE of F8-based OLEDs ⁴⁶ ,
208	no similar investigation has been reported on OLETs yet and we consider this as an
209	additional benefit of combining F8 with DAE_F. A lowering of the operating voltage
210	and enhancement of the brightness is the subject of our future studies, which clearly
211	target technological applications. It can be achieved via the careful selection of (i) the
212	light-emitting materials, which should possess high charge carrier mobility and
213	photoluminescence efficiencies, (ii) the dielectric materials, which should exhibit a
214	high gate capacitance, (iii) the optimal device structures, with a shorter channel length

and asymmetric electrodes, and (iv) a hole transport layer and/or an electron transport
layer integrated as active component¹¹⁻¹⁵.

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218 Photoswitching

The remote control via light irradiation of the electrical and optical OSOLETs 219 output was investigated. UV (315 nm) irradiation of the OSOLETs yields a significant 220 decrease of both drain current and luminance, which we attribute to ring-closing of the 221 DAEs and subsequent efficient trapping of the majority of carriers. In the case of 222 F8T2/DAE tBu bicomponent devices, the on/off ratios (the ratio of the 223 current/luminance in the initial state and after UV irradiation) in both transport current 224 and EL exceed 500 (see Figs. 2d and 2g). Such a large degree of modulation is 225 comparable with the reports of photo-programmable OLEDs and non-volatile organic 226 memories^{47,48}. A high modulation of the drain current and luminance was also observed 227 for MDMO-PPV/DAE tBu OSOLET devices (85 % decrease in current and 87 % 228 decrease in luminance, at $V_g = -120$ V, see Figs. 2e and 2h) as well as F8/DAE F 229 OSOLET devices (65 % decrease in current and 75 % decrease in luminance, at $V_g = -$ 230 100 V, see Figs. 2f and 2i). Deeper investigations into the amount of incorporated 231 photochromic molecules, energy levels matching, irradiation dose, and the thickness of 232 active film, will surely enable further improvement of the ON/OFF ratios^{22,47-48}. Further 233 irradiation with green (528 nm) light converts the DAEs back to their open form, and 234 leads to full recovery of the initial state in all three-colour OSOLETs. Under the same 235

- irradiation conditions, neat polymer based OLETs did not show any modulation ofoutput current and EL (see supplementary Fig. S7).
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It should be noted that the energy transfer from the light-emitting polymers to 239 DAEs in their closed form, favoured by the spectral overlap of the emission of the 240 polymers with the absorption of the DAEs in ring-closed form, can also contribute to 241 the modulation of the intensity of the emitted light. Indeed, a slight modulation of the 242 photoluminescence spectra was detected on the light-emitting polymer/DAE films upon 243 UV and visible irradiation (see supplementary Fig. S9). However, as the switching 244 phenomenon can be observed both on charge transport and light emission, we conclude 245 that energy transfer is not the main operating principle of OSOLETs. Nevertheless, the 246 energy transfer process might help further modulate the intensity of EL, which can be 247 used to explain the larger modulation degree of the luminance than of the current. 248

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250 The effect of DAE photoswitching on both output current and luminance of the OSOLETs was monitored over three cycles with alternative UV and visible irradiation 251 (Fig. 3). The large modulation of both maximum drain current and luminance of the 252 three-colour OSOLETs, normalized to the initial value for each measurement, is 253 reversible and the optical switching behaviour of OSOLETs is stable over several cycles. 254 Reference OLETs prepared with pristine light-emitting polymers did not show any 255 optically induced modulation, neither on drain current nor on luminance by UV or 256 visible irradiation (see supplementary Fig. S8). 257

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259	Reversibly write and erase emitting patterns More importantly, since light is used
260	as an external control of our OSOLETs, it is possible to generate emitting patterns with
261	high spatial and temporal resolution on a single transistor as demonstrated by optical
262	images of patterns created and erased in a single OSOLET (Fig. 4). In the initial
263	configuration, the F8T2/DAE_tBu OSOLET was in an all light-emitting on-state (Fig.
264	4a). Then, the device was irradiated homogenously using UV light yielding a dark state
265	(Fig. 4b). More interestingly, a well-focused green laser (532 nm) can be used to form
266	patterns on the OSOLET with the aid of a microscope. Fig. 4c shows a light-emitting
267	on-area with a pattern of 'H' shape on the same device. Subsequently, the irradiation of
268	the entire device with green light erased the patterns and the all light-emitting on-state
269	can be seen again (Fig. 4d). The second pattern can be written in the same pixel with
270	another step of UV irradiation and followed by one more laser writing step. Fig. 4f
271	exhibits a pattern image of an array of dots in the same area, demonstrating the
272	reversible and reproducible patterning of emissive features up to the micron/sub-micron
273	scale. This demonstration using an optimized combination of photoswitch and emissive
274	polymer illustrates the great potential of our approach to reversibly encode high-density
275	visual information in a single display pixel.
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277 Conclusions

We have fabricated a novel OLET device in which the transport current and electroluminescence, emitting in the range of the three primary colours, can be switched efficiently and reversibly using light as an external stimulus. The active layer in such

optically switchable organic light-emitting transistor comprises an organic light-281 emitting semiconductor and a photochromic DAE, in which the photo-tunable energy 282 levels of DAEs can either transport or trap the charge carriers, toggled by UV and 283 visible light irradiation. Such a dual external control is achieved through the 284 engineering of the energy levels of the light-emitting polymers and those of the DAEs 285 isomers. We are able to write and erase emitting patterns in a single OSOLET through 286 a non-invasive and mask-free process, with a spatial resolution of a few micrometres, 287 and a response on the microsecond time scale²². The stimuli-responsive multifunctional 288 devices proposed in this work are all readily fabricated via solution processing, thus 289 potentially transferrable to roll-to-roll compatible or ink-jet printing lines to produce 290 low-cost and flexible stimuli-responsive (nano)electronics on a large scale. Ink-jet 291 292 printing appears being a most suitable deposition method to fabricate full-colour displays by precisely positioning red, green, and blue emissive inks on each sub-pixel. 293 In principle, light emission from OLETs in any region of the visible spectrum can 294 be tuned by choosing appropriate photochromic molecules in combination with suitable 295 light-emitting polymers. Future efforts will be directed towards optically switchable 296 OLETs that can be operated at a lower driving power/voltage and yield stronger 297 brightness and a higher ON/OFF ratio. Our approach opens intriguing perspectives 298 towards the development of novel optically gated, integrated full-colour displays, 299 micro-sized light sensors, active optical memories, light controlled inverters, and logic 300 circuitries. 301 302

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423 Author contributions

L.H., X.Z., and P.S. conceived the experiments. M.H., B.M.S., M.P., and S.H. 424 425 synthesized the DAEs. L.H. did UV/visible absorption and PL measurements, and X.Z. performed AFM (the F8/DAE tBu sample by G.C.) and CV measurements. L.H. and 426 427 X.Z. designed the devices, performed the electrical experiments and carried out 428 emitting patterns writings. G.F.C., G.C., and F.C designed and built the device characterisation setup. G.F.C., G.C., and L.H. performed the quantitative OLET device 429 characterisation. All authors discussed the results and contributed to the interpretation 430 of data. L.H., X.Z., and P.S. co-wrote the paper with inputs from all co-authors. 431

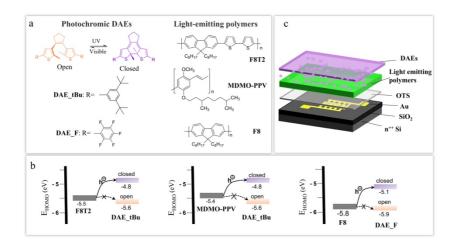
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433 Data available statement

434 The data that support the plots within this paper and other findings of this study are435 available from the corresponding author upon reasonable request.

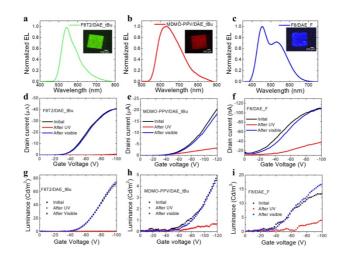
436 437 **Competing interests** The authors declare no competing financial interests 438 439 440 **Additional information** 441 Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. 442 Correspondence and requests for materials should be addressed to P.S. 443 444

445 Figure captions



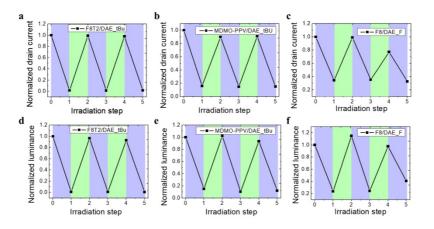
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447 Fig. 1 | Molecules, energetics, and device structure of optically switchable organic light-emitting transistors. a, Chemical structures of photochromic diarylethenes 448 (DAE tBu and DAE F) and light-emitting polymers (green: F8T2; red: MDMO-PPV; 449 blue: F8). b, Schematic illustration of the switchable charge trapping mechanism of 450 OSOLETs based on the HOMO energy levels of DAEs. As an illustrative example, for 451 F8T2, its hole transport is greatly favoured to the closed form of DAE tBu but not to 452 the open form of the latter. Thus, there is minimal trapping for the open form, while it 453 is significant for the closed form of DAE tBu. c, Structure of the OSOLETs (substrate, 454 dielectric layer, source and drain electrodes are the same for all the devices, while the 455 light-emitting polymers and DAEs are varied). 456



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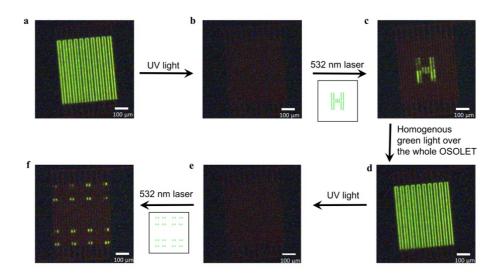
Fig. 2 | Electroluminescence spectra, optical micrographs, and optoelectronic 458 characteristics. a to c, EL spectra and emitting images of F8T2/DAE tBu, MDMO-459 PPV/DAE tBu and F8/DAE F containing OSOLETs. The scale bar in the inset is 100 460 μ m. d to f, Transfer characteristic curves of F8T2/DAE tBu OSOLET (V_d = -100 V), 461 MDMO-PPV/DAE tBu OSOLET ($V_d = -120 V$) and F8/DAE F OSOLET ($V_d = -100$ 462 V), and light-triggered current switching upon UV and visible light irradiation. g to i, 463 Luminance and light-triggered switching of the luminance in green, red and blue 464 OSOLETs upon UV and visible light irradiation. 465



466

467 Fig. 3 | Reversible modulation of optically switchable organic light-emitting
468 transistors' current and luminance during irradiation cycles. a-c Drain current and
469 d-f luminance of green, red, and blue OSOLETs over three irradiation cycles with UV
470 light (315 nm, 0.6 mW, 10 min, violet shaded areas) and visible green light (528 nm,

- $471 \quad 7 \text{ mW}, 90 \text{ s}, \text{ green shaded areas}$). All values are normalized to initial value before any
- 472 irradiation and the connecting lines serve as 'guides to the eye'.



473

Fig. 4 | Emitting pattern created and erased within a single optically switchable 474 organic light-emitting transistor. a, Optical image of a F8T2/DAE tBu OSOLET 475 device biased with $V_d = V_g = -100$ V. **b**, Optical image of a dark state in the same 476 device after UV light irradiation. c, Optical image of an 'H' shape emitting pattern from 477 the OSOLET written with a well-focused green laser (532 nm). d, Optical image of the 478 second all light-emitting on-state upon homogenous green light irradiation to erase the 479 pattern. e, Optical image of the second dark state after homogenous UV irradiation. f, 480 Optical image of an array of dots emitting patterns written by the irradiation with a 481 532 nm laser. The dark area in the pattern is due to the variation of the intensity and/or 482 focus of the laser. 483