

 Open access • Journal Article • DOI:10.1038/S41565-019-0370-9

Optically switchable organic light-emitting transistors. — [Source link](#)

[Lili Hou](#), [Xiaoyan Zhang](#), [Xiaoyan Zhang](#), [Giovanni F. Cotella](#) ...+8 more authors

Institutions: [University of Strasbourg](#), [Chalmers University of Technology](#), [London Centre for Nanotechnology](#), [Humboldt University of Berlin](#) ...+1 more institutions

Published on: 18 Feb 2019 - [Nature Nanotechnology](#) (Nat Nanotechnol)

Topics: [Nanoelectronics](#)

Related papers:

- [Flexible non-volatile optical memory thin-film transistor device with over 256 distinct levels based on an organic bicomponent blend](#)
- [Optically switchable transistor via energy-level phototuning in a bicomponent organic semiconductor](#)
- [Photochromism of Diarylethene Molecules and Crystals: Memories, Switches, and Actuators](#)
- [High-Efficiency Single-Component Organic Light-Emitting Transistors.](#)
- [Optically switchable transistors by simple incorporation of photochromic systems into small-molecule semiconducting matrices.](#)

Share this paper:    

View more about this paper here: <https://typeset.io/papers/optically-switchable-organic-light-emitting-transistors-12nj08y7y0>



HAL
open science

Optically switchable organic light-emitting transistors

Lili Hou, Xiaoyan Zhang, Giovanni F. Cotella, Giuseppe Carnicella, Martin Herder, Bernd M. Schmidt, Michael Pätzel, Stefan Hecht, Franco Cacialli, Paolo Samorì

► **To cite this version:**

Lili Hou, Xiaoyan Zhang, Giovanni F. Cotella, Giuseppe Carnicella, Martin Herder, et al.. Optically switchable organic light-emitting transistors. Nature Nanotechnology, Nature Publishing Group, 2019, 10.1038/s41565-019-0370-9 . hal-02054649

HAL Id: hal-02054649

<https://hal.archives-ouvertes.fr/hal-02054649>

Submitted on 1 Mar 2019

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

1 **Optically switchable organic light-emitting transistors**

2 Lili Hou¹, Xiaoyan Zhang^{1,4}, Giovanni F. Cotella², Giuseppe Carnicella², Martin
3 Herder³, Bernd M. Schmidt^{3,5}, Michael Pätzel³, Stefan Hecht^{3,*}, Franco Cacialli^{2,*},
4 Paolo Samori^{1,*}

5 1 Université de Strasbourg, CNRS, ISIS, 8 allée Gaspard Monge, 67000 Strasbourg, France. Email: samori@unistra.fr

6 2 Department of Physics and Astronomy (CMMP Group) and London Centre for Nanotechnology, University College London,
7 Gower Street, London WC1E 6BT, United Kingdom. Email: f.cacialli@ucl.ac.uk

8 3 Department of Chemistry & IRIS Adlershof, Humboldt-Universität zu Berlin, Brook-Taylor-Str. 2, 12489 Berlin, Germany. Email:
9 sh@chemie.hu-berlin.de

10 4 Present address: Department of Chemistry and Chemical Engineering, Chalmers University of Technology, 412 96 Gothenburg,
11 Sweden.

12 5 Present address: Heinrich Heine University Düsseldorf, Institute for Organic Chemistry and Macromolecular Chemistry,
13 Universitätsstraße 1, 40225 Düsseldorf, Germany.

14
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**
21 **solution-processed films at selected wavelengths enables the efficient and**
22 **reversible tuning of charge transport and electroluminescence simultaneously,**
23 **with a high degree of modulation (on/off ratios up to 500) in the three primary**
24 **colours. Different emitting patterns can be written and erased, through a non-**
25 **invasive and mask-free process, on a length scale of few microns in a single device,**
26 **thereby rendering this technology potentially promising for optically gated highly-**
27 **integrated full-colour displays and active optical memory.**

28
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 lasers¹⁻⁵. 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

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

47

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

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

62

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

71

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

81 with a spatial resolution down to the far-field diffraction limit, i.e. $\lambda/2NA < 1 \mu\text{m}$ (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 ($\sim 55.5 \mu\text{m}$), the present system holds particular potential of reversibly encoding high-
85 density visual information into a single pixel of a high-resolution display.

86

87 **Mechanism and device fabrication**

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

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

111

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

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

134

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

143

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

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

161

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

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 ($L = 20 \mu\text{m}$) (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).

176

177

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

191

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

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 $V_g = -80$ 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 (1.2×10^{-2}
206 %). Although the controlled formation of the β -phase of F8 *via* dipping in solvent/non-
207 solvent mixtures has already been exploited to improve the EQE of F8-based OLEDs⁴⁶,
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

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

217

218 **Photoswitching**

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

236 irradiation conditions, neat polymer based OLETs did not show any modulation of
237 output current and EL (see supplementary Fig. S7).

238

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

249

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

258

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.

276

277 **Conclusions**

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

281 optically switchable organic light-emitting transistor comprises an organic light-
282 emitting semiconductor and a photochromic DAE, in which the photo-tunable energy
283 levels of DAEs can either transport or trap the charge carriers, toggled by UV and
284 visible light irradiation. Such a dual external control is achieved through the
285 engineering of the energy levels of the light-emitting polymers and those of the DAEs
286 isomers. We are able to write and erase emitting patterns in a single OSOLET through
287 a non-invasive and mask-free process, with a spatial resolution of a few micrometres,
288 and a response on the microsecond time scale²². The stimuli-responsive multifunctional
289 devices proposed in this work are all readily fabricated *via* solution processing, thus
290 potentially transferrable to roll-to-roll compatible or ink-jet printing lines to produce
291 low-cost and flexible stimuli-responsive (nano)electronics on a large scale. Ink-jet
292 printing appears being a most suitable deposition method to fabricate full-colour
293 displays by precisely positioning red, green, and blue emissive inks on each sub-pixel.

294 In principle, light emission from OLETs in any region of the visible spectrum can
295 be tuned by choosing appropriate photochromic molecules in combination with suitable
296 light-emitting polymers. Future efforts will be directed towards optically switchable
297 OLETs that can be operated at a lower driving power/voltage and yield stronger
298 brightness and a higher ON/OFF ratio. Our approach opens intriguing perspectives
299 towards the development of novel optically gated, integrated full-colour displays,
300 micro-sized light sensors, active optical memories, light controlled inverters, and logic
301 circuitries.

302
303

304 References

- 305 1 Muccini, M. A bright future for organic field-effect transistors. *Nature Mater.* **5**, 605–613 (2006).
- 306 2 Cicoira, F. & Santato, C. Organic Light Emitting Field Effect Transistors: Advances and
307 Perspectives. *Adv. Funct. Mater.* **17**, 3421-3434 (2007).
- 308 3 Santato, C., Cicoira, F. & Martel, R. Spotlight on organic transistors. *Nature Photon.* **5**, 392–
309 393 (2011).
- 310 4 Zhang, C., Chen, P. & Hu, W. Organic light-emitting transistors: materials, device
311 configurations, and operations. *Small* **12**, 1252-1294 (2016).
- 312 5 Zaumseil, J. & Sirringhaus, H. Electron and ambipolar transport in organic field-effect
313 transistors. *Chem. Rev.* **107**, 1296-1323 (2007).
- 314 6 Zaumseil, J., Friend, R. H. & Sirringhaus, H. Spatial control of the recombination zone in an
315 ambipolar light-emitting organic transistor. *Nature Mater.* **5**, 69–74 (2005).
- 316 7 Hsu, B. B. Y. *et al.* Control of efficiency, brightness, and recombination zone in light-emitting
317 field effect transistors. *Adv. Mater.* **24**, 1171-1175 (2012).
- 318 8 Swensen, J. S., Soci, C. & Heeger, A. J. Light emission from an ambipolar semiconducting
319 polymer field-effect transistor. *Appl. Phys. Lett.* **87**, 253511 (2005).
- 320 9 Bisri, S. Z. *et al.* High mobility and luminescent efficiency in organic single-crystal light-
321 emitting transistors. *Adv. Funct. Mater.* **19**, 1728–1735 (2009).
- 322 10 Capelli, R. *et al.* Interface functionalities in multilayer stack organic light emitting transistors
323 (OLETs). *Adv. Funct. Mater.* **24**, 5603-5613 (2014).
- 324 11 Capelli, R. *et al.* Organic light-emitting transistors with an efficiency that outperforms the
325 equivalent light-emitting diodes. *Nature Mater.* **9**, 496–503 (2010).
- 326 12 McCarthy, M. A. *et al.* Low-voltage, low-power, organic light-emitting transistors for active
327 matrix displays. *Science* **332**, 570–573 (2011).
- 328 13 Zaumseil, J., Donley, C. L., Kim, J. S., Friend, R. H. & Sirringhaus, H. Efficient top-gate,
329 ambipolar, light-emitting field-effect transistors based on a green-light-emitting polyfluorene.
330 *Adv. Mater.* **18**, 2708–2712 (2006).
- 331 14 Gwinner, M. C. *et al.* Highly efficient single-layer polymer ambipolar light-emitting field-effect
332 transistors. *Adv. Mater.* **24**, 2728–2734 (2012).
- 333 15 Ullah, M. *et al.* Simultaneous enhancement of brightness, efficiency, and switching in RGB
334 organic light emitting transistors. *Adv. Mater.* **25**, 6213–6218 (2013).
- 335 16 Park, S. K. *et al.* Highly luminescent 2D-type slab crystals based on a molecular charge-transfer
336 complex as promising organic light-emitting transistor materials. *Adv. Mater.* **29**, 1701346
337 (2017).
- 338 17 Yang, Y., da Costa, R. C., Fuchter, M. J. & Campbell, A. J. Circularly polarized light detection
339 by a chiral organic semiconductor transistor. *Nature Photon.* **7**, 634–638 (2013).
- 340 18 Kim, Y. L. *et al.* Voltage-switchable photocurrents in single-walled carbon nanotube–silicon
341 junctions for analog and digital optoelectronics. *Nature Photon.* **8**, 239–243 (2014).
- 342 19 Zhang, Y. J., Oka, T., Suzuki, R., Ye, J. T. & Iwasa, Y. Electrically switchable chiral light-
343 emitting transistor. *Science* **344**, 725–728 (2014).
- 344 20 Wang, X. *et al.* A spectrally tunable all-graphene-based flexible field-effect light-emitting
345 device. *Nature Commun.* **6**, 7767 (2015).
- 346 21 Zhang, X. Y., Hou, L. L. & Samorì, P. Coupling carbon nanomaterials with photochromic
347 molecules for the generation of optically responsive materials. *Nature Commun.* **7**, 11128 (2016).

348 22 Orgiu, E. *et al.* Optically switchable transistor via energy-level phototuning in a bicomponent
349 organic semiconductor. *Nature Chem.* **4**, 675–679 (2012).

350 23 Gemayel, M. E. *et al.* Optically switchable transistors by simple incorporation of photochromic
351 systems into small-molecule semiconducting matrices. *Nature Commun.* **6**, 6330 (2015).

352 24 Borjesson, K. *et al.* Optically switchable transistors comprising a hybrid photochromic
353 molecule/n-type organic active layer. *J. Mater. Chem. C* **3**, 4156–4161 (2015).

354 25 Irie, M. & Mohri, M. Thermally irreversible photochromic systems. Reversible
355 photocyclization of diarylethene derivatives. *J. Org. Chem.* **53**, 803–808 (1988).

356 26 Irie, M., Fukaminato, T., Matsuda, K. & Kobatake, S. Photochromism of diarylethene molecules
357 and crystals: memories, switches, and actuators. *Chem. Rev.* **114**, 12174–12277 (2014).

358 27 Hou, L., Zhang, X., Pijper, T. C., Browne, W. R. & Feringa, B. L. Reversible photochemical
359 control of singlet oxygen generation using diarylethene photochromic switches. *J. Am. Chem.*
360 *Soc.* **136**, 910–913 (2014).

361 28 Leydecker, T. *et al.* Flexible non-volatile optical memory thin-film transistor device with over
362 256 distinct levels based on an organic bicomponent blend. *Nature Nanotech.* **11**, 769–775
363 (2016).

364 29 Herder, M. *et al.* Improving the fatigue resistance of diarylethene switches. *J. Am. Chem. Soc.*
365 **137**, 2738–2747 (2015).

366 30 Herder, M. *et al.* Light-controlled reversible modulation of frontier molecular orbital energy
367 levels in trifluoromethylated diarylethenes. *Chem. Eur. J.* **23**, 3743–3754 (2017).

368 31 Salleo, A., Chabinyo, M. L., Yang, M. S. & Street, R. A. Polymer thin-film transistors with
369 chemically modified dielectric interfaces. *Appl. Phys. Lett.* **81**, 4383–4385 (2002).

370 32 Ito, Y. *et al.* Crystalline ultrasoft self-assembled monolayers of alkylsilanes for organic field-
371 effect transistors. *J. Am. Chem. Soc.* **131**, 9396–9404 (2009).

372 33 Lee, T.-W. & Park, O. O. The effect of different heat treatments on the luminescence efficiency
373 of polymer light-emitting diodes. *Adv. Mater.* **12**, 801–804 (2000).

374 34 Grell, M., Bradley, D. D. C., Inbasekaran, M. & Woo, E. P. A glass-forming conjugated main-
375 chain liquid crystal polymer for polarized electroluminescence applications. *Adv. Mater.* **9**, 798–
376 802 (1997).

377 35 Perevedentsev, A., Chander, N., Kim, J. S. & Bradley, D. D. C. Spectroscopic properties of
378 poly(9,9-dioctylfluorene) thin films possessing varied fractions of β -phase chain segments:
379 enhanced photoluminescence efficiency via conformation structuring. *J. Polym. Sci. Pol. Phys.*
380 **54**, 1995–2006 (2016).

381 36 Caruso, M. E., Lattante, S., Cingolani, R. & Anni, M. Microscopic investigation of the poly(9,9-
382 dioctylfluorene) photoluminescence dependence on the deposition conditions by confocal laser
383 microscopy. *Appl. Phys. Lett.* **88**, 181906 (2006).

384 37 Lim, S. F. *et al.* Suppression of green emission in a new class of blue-emitting PF copolymers
385 with twisted biphenyl moieties. *Adv. Funct. Mater.* **15**, 981–988 (2005).

386 38 List, E. J. W., Guentner, R., Scanducci de Freitas, P. & Scherf, U. The effect of keto defect sites
387 on the emission properties of polyfluorene-type materials. *Adv. Mater.* **14**, 374–378 (2002).

388 39 Honmou, Y. *et al.* Single-molecule electroluminescence and photoluminescence of polyfluorene
389 unveils the photophysics behind the green emission band. *Nature Commun.* **5**, 4666 (2014).

390 40 Gong, X. *et al.* Stabilized blue emission from polyfluorene-based light-emitting diodes:
391 elimination of fluorenone defects. *Adv. Funct. Mater.* **13**, 325–330 (2003).

- 392 41 Hepp, A. *et al.* Light-emitting field-effect transistor based on a tetracene thin film. *Phys. Rev.*
393 *Lett.* **91**, 157406 (2003).
- 394 42 Santato, C. *et al.* Tetracene-based organic light-emitting transistors: optoelectronic properties
395 and electron injection mechanism. *Synth. Met.* **146**, 329–334 (2004).
- 396 43 Roelofs, W. S. C., Adriaans, W. H., Janssen, R. A. J., Kemerink, M. & Leeuw, D. M. d. Light
397 emission in the unipolar regime of ambipolar organic field-effect transistors. *Adv. Funct. Mater.*
398 **23**, 4133–4139 (2013).
- 399 44 Redecker, M., Bradley, D. D. C., Inbasekaran, M. & Woo, E. P. Mobility enhancement through
400 homogeneous nematic alignment of a liquid-crystalline polyfluorene. *Appl. Phys. Lett.* **74**,
401 1400–1402 (1999).
- 402 45 Hsu, B. B. Y. *et al.* Ordered polymer nanofibers enhance output brightness in bilayer light-
403 emitting field-effect transistors. *ACS Nano* **7**, 2344–2351 (2013).
- 404 46 Lu, H. H., Liu, C. Y., Chang, C. H. & Chen, S. A. Self-dopant formation in poly(9,9-di-n-
405 octylfluorene) via a dipping method for efficient and stable pure-blue electroluminescence. *Adv.*
406 *Mater.* **19**, 2574–2579 (2007).
- 407 47 Zacharias, P., Gather, M. C., Köhnen, A., Rehmann, N. & Meerholz, K. Photoprogrammable
408 organic light-emitting diodes. *Angew. Chem. Int. Ed.* **48**, 4038–4041 (2009).
- 409 48 Scott, J. C. & Bozano, L. D. Nonvolatile memory elements based on organic materials. *Adv.*
410 *Mater.* **19**, 1452–1463 (2007).

411

412 **Acknowledgement**

413 We acknowledge funding from the European Commission through the Marie
414 Sklodowska-Curie ITN project iSwitch (GA-642196), the Marie Sklodowska-Curie
415 ITN project SYNCHRONICS (GA-643238), the ERC projects SUPRAFUNCTION
416 (GA-257305) and LIGHT4FUNCTION (GA-308117), the Agence Nationale de la
417 Recherche through the Labex project CSC (ANR-10-LABX-0026 CSC) within the
418 Investissement d’Avenir program (ANR-10-120 IDEX-0002-02), and the International
419 Center for Frontier Research in Chemistry (icFRC) as well as the German Research
420 Foundation (via SFB 765 and SFB 951). FC is a Royal Society Wolfson Research Merit
421 Award holder.

422

423 **Author contributions**

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

432

433 **Data available statement**

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

436

437 Competing interests

438 The authors declare no competing financial interests

439

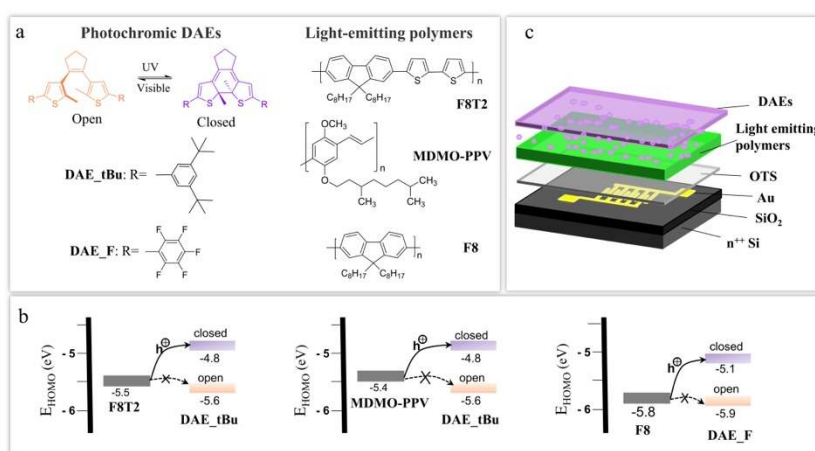
440 Additional information

441 Supplementary information is available in the online version of the paper. Reprints and
442 permissions information is available online at www.nature.com/reprints.

443 Correspondence and requests for materials should be addressed to P.S.

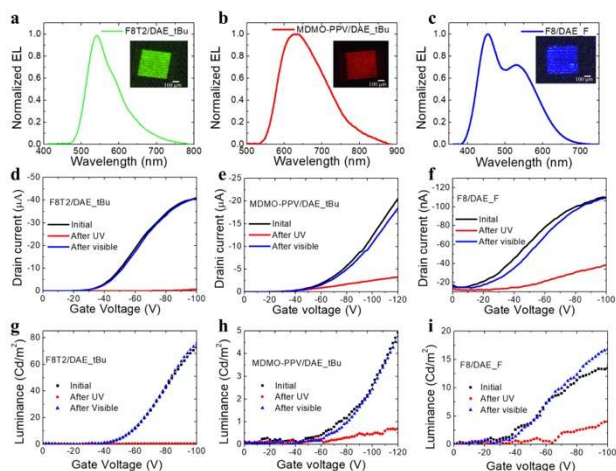
444

445 Figure captions



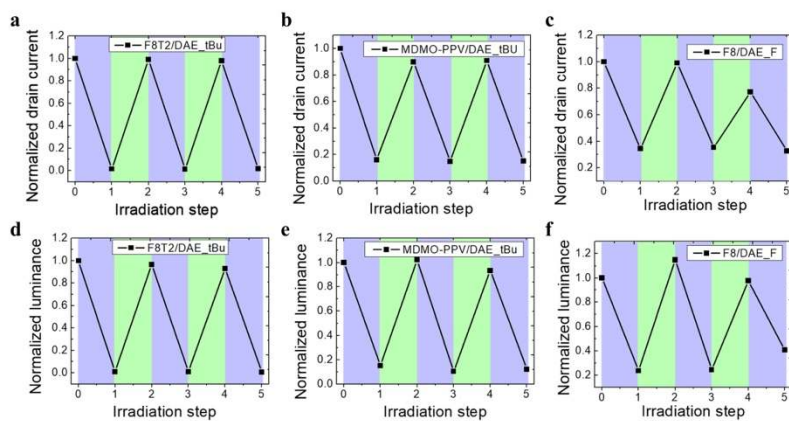
446

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



457

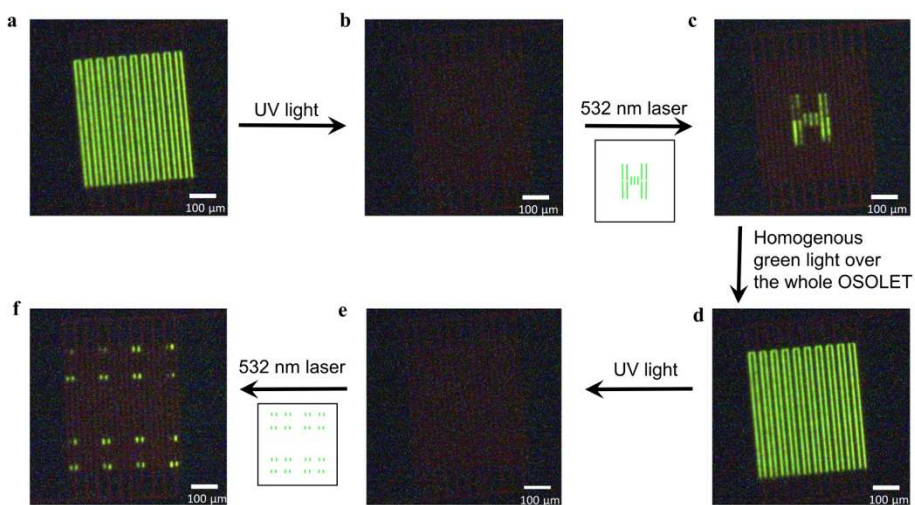
458 **Fig. 2 | Electroluminescence spectra, optical micrographs, and optoelectronic**
 459 **characteristics. a to c**, EL spectra and emitting images of F8T2/DAE_tBu, MDMO-
 460 PPV/DAE_tBu and F8/DAE_F containing OSOLETs. The scale bar in the inset is 100
 461 μm . **d to f**, Transfer characteristic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



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 7 mW, 90 s, 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

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