

## Supporting Information

for *Adv. Electron. Mater.*, DOI: 10.1002/aelm.202200293

X-ray Detectors With Ultrahigh Sensitivity Employing  
High Performance Transistors Based on a Fully Organic  
Small Molecule Semiconductor/Polymer Blend Active  
Layer

*Adrián Tamayo, Ilaria Fratelli, Andrea Ciavatti, Carme  
Martínez-Domingo, Paolo Branchini, Elisabetta  
Colantoni, Stefania De Rosa, Luca Tortora, Adriano  
Contillo, Raul Santiago, Stefan T. Bromley, Beatrice  
Fraboni,\* Marta Mas-Torrent,\* and Laura Basiricò*

## Supporting Information

**X-ray detectors with ultrahigh sensitivity employing high performance transistors based on a fully organic small molecule semiconductor/polymer blend active layer**

*Adrián Tamayo,<sup>1</sup> Ilaria Fratelli,<sup>2</sup> Andrea Ciavatti,<sup>2</sup> Carme Martínez-Domingo,<sup>1</sup> Paolo Branchini,<sup>3,4</sup> Elisabetta Colantoni,<sup>3,4,5</sup> Stefania De Rosa,<sup>3,4</sup> Luca Tortora,<sup>3,4,6</sup> Adriano Contillo,<sup>7</sup> Raul Santiago,<sup>8</sup> Stefan T. Bromley,<sup>8,9</sup> Beatrice Fraboni,<sup>2,\*</sup> Marta Mas-Torrent,<sup>1,\*</sup> Laura Basiricò<sup>2</sup>*

<sup>1</sup> Institut de Ciència de Materials de Barcelona (ICMAB-CSIC) and Networking Research Center on Bioengineering, Biomaterials and Nanomedicine (CIBER-BBN), Campus de la Universitat Autònoma de Barcelona, Cerdanyola, E-08193 Barcelona, Spain.

<sup>2</sup> Department of Physics and Astronomy, University of Bologna and National Institute for Nuclear Physics - INFN section of Bologna, Viale Berti Pichat 6/2, 40127 Bologna, Italy.

<sup>3</sup> Surface Analysis Laboratory INFN Roma Tre, Via della Vasca Navale 84, 00146 Rome, Italy.

<sup>4</sup> INFN, Roma Tre, via della Vasca Navale 84, Rome, Italy.

<sup>5</sup> Department of Mathematics and Physics, Roma Tre University, via della Vasca Navale 84, Rome, Italy.

<sup>6</sup> Department of Sciences, Roma Tre University, Via della Vasca Navale 84, Rome, Italy.

<sup>7</sup> Elettra-Sincrotrone Trieste, Area Science Park Strada Statale 14, km 163.5, 34149 Basovizza, Trieste, Italy

<sup>8</sup> Departament de Ciència de Materials i Química Física & Institut de Química Teòrica i Computacional (IQTUB), Universitat de Barcelona, Barcelona, Spain.

<sup>9</sup> Institució Catalana de Recerca i Estudis Avançats (ICREA), Barcelona, Spain

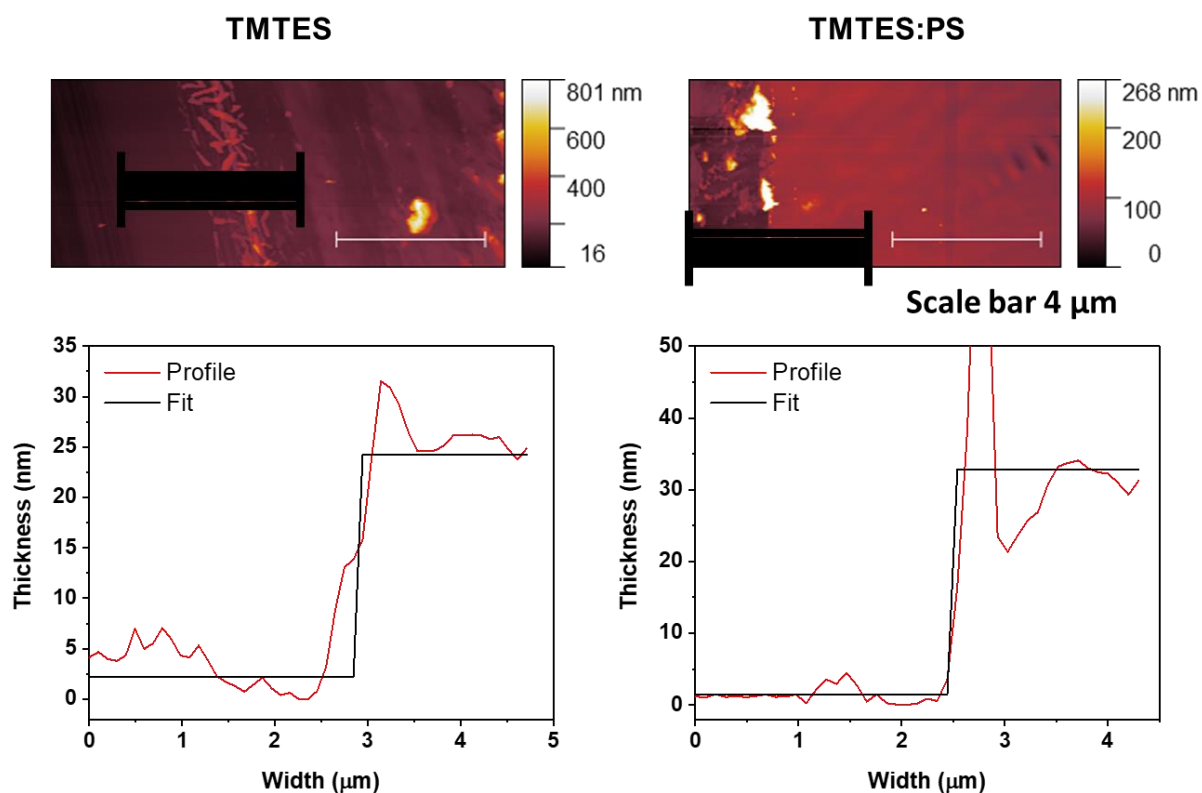
**Table S1.** Electrical parameters of OFETs based on films of TMTES and TMTES:PS deposited by BAMS. The parameters are calculated for OFETs with the conducting channel parallel ( $\parallel$ ) and perpendicular ( $\perp$ ) to the coating direction.

<i>Ink formulation</i>	<b>Speed (mm s<sup>-1</sup>)</b>		<b>Mobility (cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>)</b>	<b>V<sub>TH</sub> (V)</b>	<b>On/Off</b>	<b><math>\mu^{\max}</math> (cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>)</b>
<i>TMTES</i>	1	$\parallel$	$(1.1 \pm 0.3) \cdot 10^{-1}$	$(30 \pm 10)$	$10^3$	$0.19 \pm 0.2$
		$\perp$	$(9 \pm 4) \cdot 10^{-2}$	$(40 \pm 10)$	$10^3$	$0.11 \pm 0.2$
	10	$\parallel$	$(1.0 \pm 0.3) \cdot 10^{-1}$	$(27 \pm 9)$	$10^3$	$0.22 \pm 0.3$
		$\perp$	$(7.4 \pm 1.9) \cdot 10^{-2}$	$(34 \pm 9)$	$10^3$	$0.10 \pm 0.2$
<i>TMTES:PS</i> (4:1) <i>PS</i> <sub>10K</sub>	1	$\parallel$	$1.2 \pm 0.2$	$2 \pm 2$	$10^4$	$1.4 \pm 0.2$
		$\perp$	$0.2 \pm 0.1$	$4 \pm 3$	$10^4$	$0.4 \pm 0.2$
	10	$\parallel$	$1.3 \pm 0.3$	$1 \pm 1$	$10^4$	$1.6 \pm 0.2$
		$\perp$	$0.7 \pm 0.3$	$-0.1 \pm 0.3$	$10^4$	$0.9 \pm 0.2$
<i>TMTES:PS</i> (4:1) <i>PS</i> <sub>280K</sub>	1	$\parallel$	$0.7 \pm 0.1$	$-(2 \pm 1)$	$10^4$	$0.9 \pm 0.2$
		$\perp$	$0.4 \pm 0.3$	$-(2.6 \pm 0.7)$	$10^4$	$0.7 \pm 0.2$
	10	$\parallel$	$1.7 \pm 0.4$	$-(0.7 \pm 0.2)$	$10^4$	$2.3 \pm 0.3$
		$\perp$	$1.0 \pm 0.2$	$-(0.2 \pm 0.6)$	$10^4$	$1.5 \pm 0.2$
<b><i>TMTES:PS</i></b> <b>(2:1)</b> <b><i>PS</i><sub>280K</sub></b>	1	$\parallel$	$2.4 \pm 0.4$	$-(0.53 \pm 0.17)$	$10^5$	$3.6 \pm 0.4$
		$\perp$	$2.0 \pm 0.4$	$-(0.45 \pm 0.15)$	$10^5$	$2.5 \pm 0.3$
	10	$\parallel$	<b><math>2.6 \pm 0.6</math></b>	<b><math>-(1.1 \pm 0.2)</math></b>	<b><math>10^5</math></b>	<b><math>3.1 \pm 0.2</math></b>
		$\perp$	<b><math>2.1 \pm 0.6</math></b>	<b><math>-(1.00 \pm 0.11)</math></b>	<b><math>10^5</math></b>	<b><math>2.8 \pm 0.3</math></b>
<i>TMTES:PS</i> (1:2) <i>PS</i> <sub>280K</sub>	1	$\parallel$	$1.1 \pm 0.2$	$-(0.4 \pm 0.8)$	$10^4$	$1.6 \pm 0.2$
		$\perp$	$1.0 \pm 0.3$	$-(0.4 \pm 0.3)$	$10^4$	$1.4 \pm 0.3$
	10	$\parallel$	$0.9 \pm 0.2$	$-(1.0 \pm 0.2)$	$10^4$	$1.4 \pm 0.2$
		$\perp$	$0.7 \pm 0.3$	$-(0.9 \pm 0.3)$	$10^4$	$1.0 \pm 0.2$

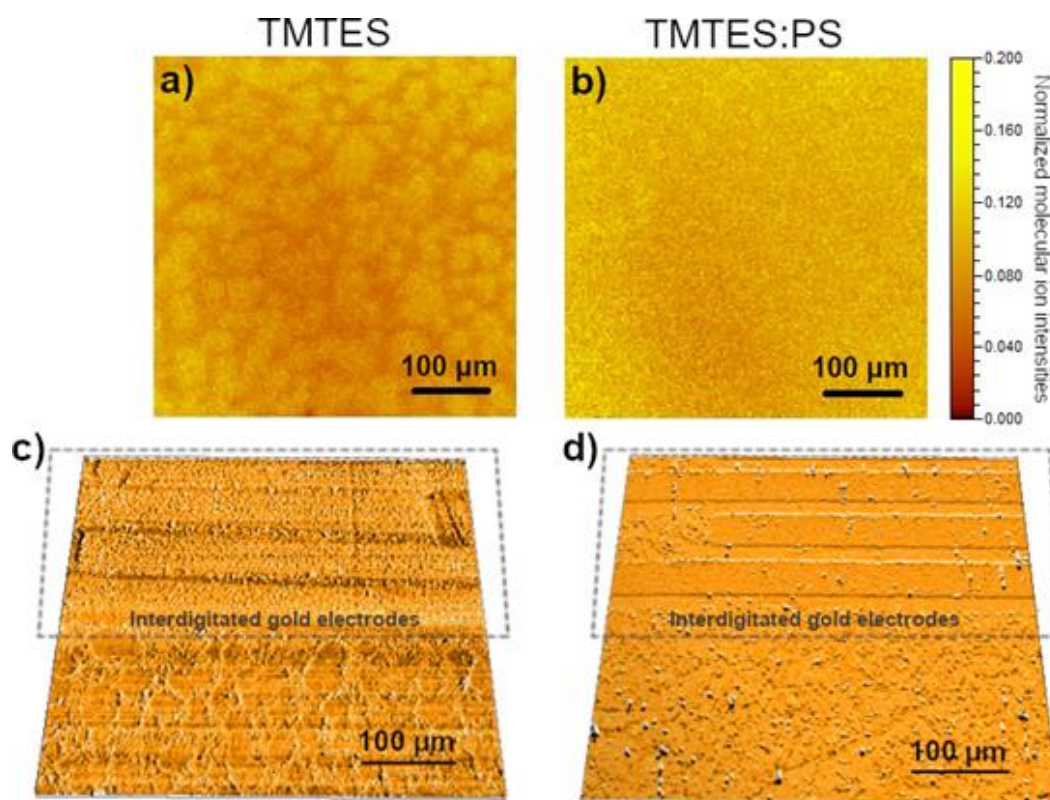
**Table S2.** Comparison of our results with the electrical transport parameters of solution processed OFETs based on TMTES reported in literature.

OSC Formulation	Scalable to Roll-to-Roll (Deposition Technique)	Binder	$\mu$ (cm <sup>2</sup> ·V <sup>-1</sup> ·s <sup>-1</sup> )	V <sub>TH</sub> (V)	Ref
2.0 wt % CB (2:1) PS	YES (Solution Shearing)	YES	2.6±0.6	-1	This work
2.0 wt % CB	YES (Solution Shearing)	NO	0.10±0.03	~30	This work
0.5 wt % TL	NO (Drop casting)	NO	1.3±0.4	NA	1
1.2 wt % TE (1:2) 4-iPrCN-TAA/C8-Flu (70:30)	NO (Spin coating)	YES	4.3±0.3	~10	2
TE	NO (Spin coating)	NO	2.6-3.5	~40	3
1wt % TE	NO (Drop casting)	NO	0.3	~0	4
1 wt% TE (1:1) (iPVN)	NO (Drop casting)	YES	0.07	~0	4
n.r.	NO (Spin coating)	NO	1.9		5

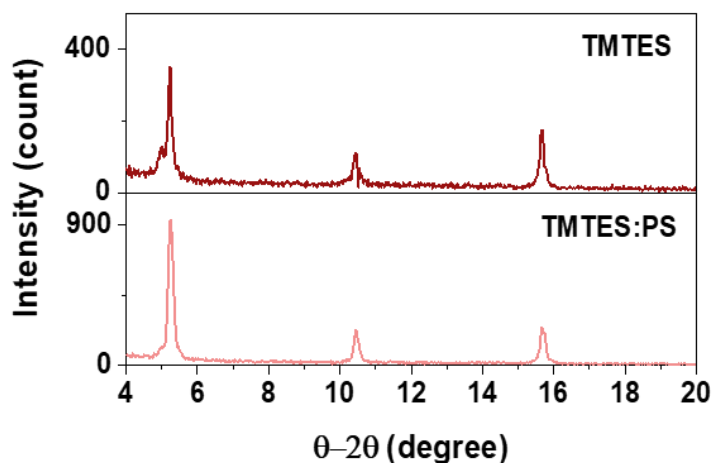
(CB) chlorobenzene, (TL) toluene, (TE) tetralin, (iPVN) isotactic poly(a-vinyl naphthalene and (4-iPrCN-TAA/C8-Flu) 4-isopropylcyano triarylamine/n-octyl Fluorene, 70/30 copolymer. n.r.: not reported.



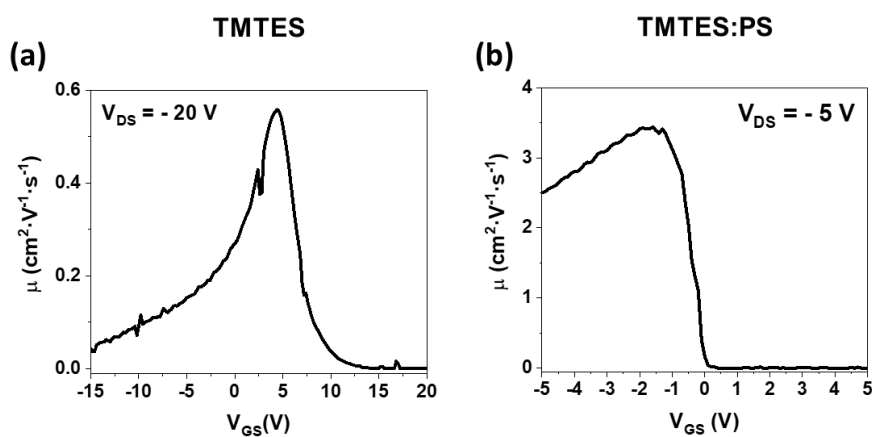
**Figure S1.** Topographic AFM images (top) and depth profiles (bottom) for thickness estimation of the TMTES and TMTES:PS thin films.



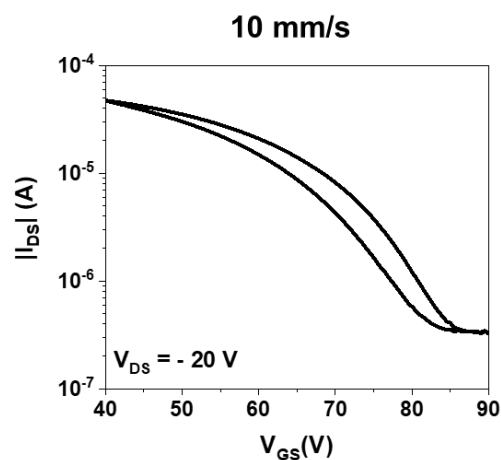
**Figure S2.** ToF-SIMS 2D surface chemical maps of TMTES and TMTES:PS thin films deposited by BAMS at high coating speed. Normalized (to total counts) sum of  $\text{Si}_2\text{C}_{42}\text{H}_{50}^+$  ( $m/z = 610.34$ ),  $\text{Si}_2^{13}\text{CC}_{41}\text{H}_{50}^+$  ( $m/z = 611.35$ ),  $\text{Si}_2^{13}\text{C}_2\text{C}_{40}\text{H}_{50}^+$  ( $m/z = 612.35$ ),  $^{30}\text{SiSi}^{13}\text{CC}_{41}\text{H}_{50}^+$  ( $m/z=613.34$ ),  $^{30}\text{SiSi}^{13}\text{C}_2\text{C}_{40}\text{H}_{50}^+$  ( $m/z = 614.35$ ), and  $^{30}\text{SiSi}^{13}\text{C}_3\text{C}_{39}\text{H}_{50}^+$  ( $m/z = 615.35$ ) secondary ion signals from (a) TMTES and (b) TMTES:PS surface acquired outside the interdigitated electrodes. 3D surface height profiles maps of c) TMTES and d) TMTES:PS films with the interdigitated gold electrodes.



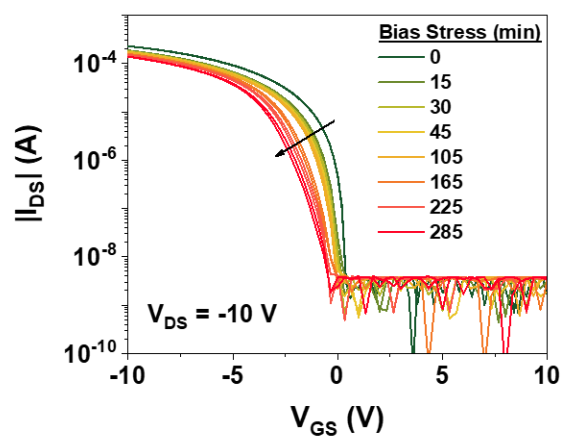
**Figure S3.** X-ray diffractograms of TMTES and TMTES:PS thin films deposited by BAMS.



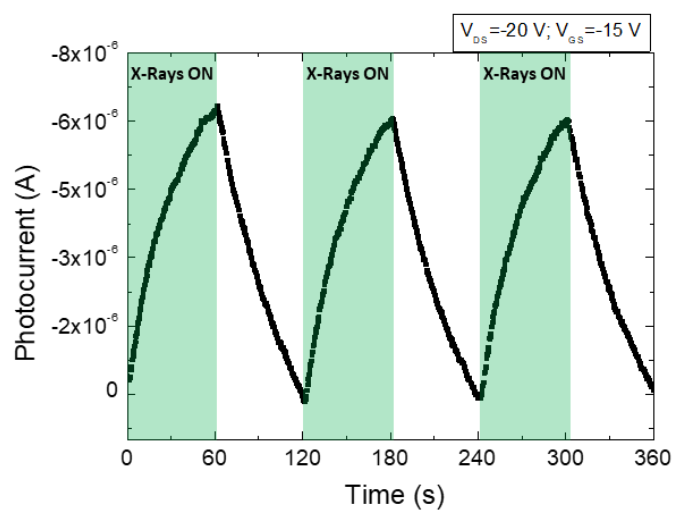
**Figure S4.** Dependence of charge carrier mobility on the applied gate voltage for OFETs based on (a) TMTES and (b) TMTES:PS.



**Figure S5.** Transfer characteristics in saturation regime of TMTES OFETs measured 90 days after their fabrication

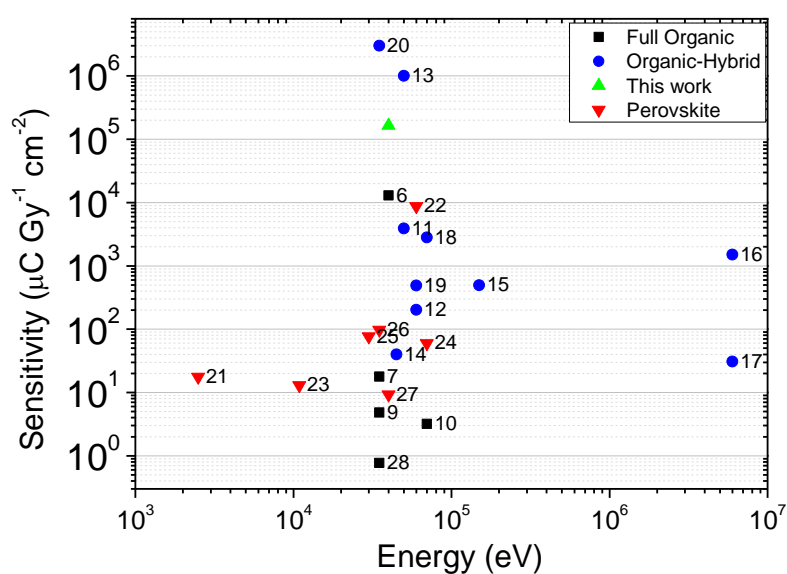


**Figure S6.** Consecutive transfer characteristics of the films TMTES:PS 2:1 under bias stress ( $V_{GS} = -10$  V and  $V_{DS} = -1$  V).



**Figure S7.** X-ray induced photocurrent response of a TMTES:PS BAMS-coated device upon three on/off switching cycles (green areas correspond to time windows of 60 s) employing a dose rate of  $9.8 \text{ mGy s}^{-1}$ .





**Figure S8.** Comparison of the sensitivity values per unit area achieved in this work (green triangle), with those reported at the state of the art for thin-film detectors based on perovskite (red triangles), organic-hybrid (blue circles), and full-organic (black squares) active layers.<sup>[6-28]</sup>

**Bibliography**

- [1] G. R. Llorente, M. B. Dufourg-Madec, D. J. Crouch, R. G. Pritchard, S. Ogier, S. G. Yeates, *Chem. Commun.* 2009, 3059.
- [2] K. L. McCall, S. R. Rutter, E. L. Bone, N. D. Forrest, J. S. Bissett, J. D. E. Jones, M. J. Simms, A. J. Page, R. Fisher, B. A. Brown, S. D. Ogier, *Adv. Funct. Mater.* 2014, 24, 3067.
- [3] J. F. Chang, T. Sakanoue, Y. Olivier, T. Uemura, M. B. Dufourg-Madec, S. G. Yeates, J. Cornil, J. Takeya, A. Troisi, H. Sirringhaus, *Phys. Rev. Lett.* 2011, 107, 066601.
- [4] M. M. Ibrahim, A. C. MacIel, C. P. Watson, M. B. Madec, S. G. Yeates, D. M. Taylor, *Org. Electron.* 2010, 11, 1234.
- [5] A. Y. B. Meneau, Y. Olivier, T. Backlund, M. James, D. W. Breiby, J. W. Andreasen, H. Sirringhaus, *Adv. Funct. Mater.* 2016, 26, 2326.
- [6] I. Temiño, L. Basiricò, I. Fratelli, A. Tamayo, A. Ciavatti, M. Mas-Torrent, B. Fraboni, *Nat. Commun.* 2020, 11, 2136.
- [7] A. Ciavatti, L. Basiricò, I. Fratelli, S. Lai, P. Cosseddu, A. Bonfiglio, J. E. Anthony, B. Fraboni, *Adv. Funct. Mater.* 2019, 29, 1806119.
- [8] A. M. Zeidell, T. Ren, D. S. Filston, H. F. Iqbal, E. Holland, J. D. Bourland, J. E. Anthony, O. D. Jurchescu, *Adv. Sci.* 2020, 7, 2001522.
- [9] S. Lai, P. Cosseddu, L. Basiricò, A. Ciavatti, B. Fraboni, A. Bonfiglio, *Adv. Electron. Mater.* 2017, 3, 1600409.

- [10] L. Basiricò, A. Ciavatti, I. Fratelli, D. Dreossi, G. Tromba, S. Lai, P. Cosseddu, A. Bonfiglio, F. Mariotti, C. Dalla Val, V. Bellucci, J. E. Anthony, B. Fraboni, *Front. Phys.* 2020, 8, 10.3389/fphy.2020.00013.
- [11] H. M. Thirimanne, K. D. G. I. Jayawardena, A. J. Parnell, R. M. I. Bandara, A. Karalasingam, S. Pani, J. E. Huerdler, D. G. Lidzey, S. F. Tedde, A. Nisbet, C. A. Mills, S. R. P. Silva, *Nat. Commun.* 2018, 9, 2926.
- [12] L. Mao, Y. Li, H. Chen, L. Yu, J. Zhang, *Nanomaterials* 2021, 11, 1832.
- [13] Y. Gao, Y. Ge, X. Wang, J. Liu, W. Liu, Y. Cao, K. Gu, Z. Guo, Y. M. Wei, N. Zhou, D. Yu, H. Meng, X. F. Yu, H. Zheng, W. Huang, J. Li, *Adv. Mater.* 2021, 33, 2101717.
- [14] H. Li, X. Shan, J. N. Neu, T. Geske, M. Davis, P. Mao, K. Xiao, T. Siegrist, Z. Yu, J. Mater. Chem. C 2018, 6, 11961.
- [15] A. Ciavatti, R. Sorrentino, L. Basiricò, B. Passarella, M. Caironi, A. Petrozza, B. Fraboni, *Adv. Funct. Mater.* 2021, 31, 2009072.
- [16] M. P. A. Nanayakkara, L. Matjačić, S. Wood, F. Richheimer, F. A. Castro, S. Jenatsch, S. Züfle, R. Kilbride, A. J. Parnell, M. G. Masteghin, H. M. Thirimanne, A. Nisbet, K. D. G. I. Jayawardena, S. R. P. Silva, *Adv. Funct. Mater.* 2021, 31, 2008482.
- [17] H. M. Thirimanne, K. D. G. I. Jayawardena, A. Nisbet, Y. Shen, R. M. I. Bandara, C. A. Mills, G. Shao, S. R. P. Silva, *IEEE Trans. Nucl. Sci.* 2020, 67, 2238.
- [18] K. D. G. I. Jayawardena, H. M. Thirimanne, S. F. Tedde, J. E. Huerdler, A. J. Parnell, R. M. I. Bandara, C. A. Mills, S. R. P. Silva, *ACS Nano* 2019, 13, 6973.
- [19] J. Peng, K. Ye, Y. Xu, L. Cui, R. Li, H. Peng, Q. Lin, *Sensors Actuators, A Phys.* 2020, 312, 112132.

- [20] A. Ciavatti, T. Cramer, M. Carroli, L. Basiricò, R. Fuhrer, D. M. De Leeuw, B. Fraboni, *Appl. Phys. Lett.* 2017, 111, 183301.
- [21] J. Liu, B. Shabbir, C. Wang, T. Wan, Q. Ou, P. Yu, A. Tadich, X. Jiao, D. Chu, D. Qi, D. Li, R. Kan, Y. Huang, Y. Dong, J. Jasieniak, Y. Zhang, Q. Bao, *Adv. Mater.* 2019, 31, 1901644.
- [22] J. Zhao, L. Zhao, Y. Deng, X. Xiao, Z. Ni, S. Xu, J. Huang, *Nat. Photonics* 2020, 14, 612.
- [23] H. Tsai, F. Liu, S. Shrestha, K. Fernando, S. Tretiak, B. Scott, D. T. Vo, J. Strzalka, W. Nie, *Sci. Adv.* 2020, 6, eaay0815.
- [24] H. Mescher, F. Schackmar, H. Eggers, T. Abzieher, M. Zuber, E. Hamann, T. Baumbach, B. S. Richards, G. Hernandez-Sosa, U. W. Paetzold, U. Lemmer, *ACS Appl. Mater. Interfaces* 2020, 12, 15774.
- [25] J. Guo, Y. Xu, W. Yang, B. Xiao, Q. Sun, X. Zhang, B. Zhang, M. Zhu, W. Jie, *ACS Appl. Mater. Interfaces* 2021, 13, 23928.
- [26] L. Basiricò, S. P. Senanayak, A. Ciavatti, M. Abdi-Jalebi, B. Fraboni, H. Sirringhaus, *Adv. Funct. Mater.* 2019, 29, 1902346.
- [27] S. Demchyshyn, M. Verdi, L. Basiricò, A. Ciavatti, B. Hailegnaw, D. Cavalcoli, M. C. Scharber, N. S. Sariciftci, M. Kaltenbrunner, B. Fraboni, *Adv. Sci.* 2020, 7, 2002586.
- [28] L. Basiricò, A. Ciavatti, T. Cramer, P. Cosseddu, A. Bonfiglio, B. Fraboni, *Nat. Commun.* **2016**, 7, 13063.