

Supporting Information (SI)
**Influence of Hole Transport layers on Internal Absorption,
Charge Recombination and Collection in HC(NH₂)₂PbI₃
Perovskite Solar Cells**

Yang Liu^a, Hao Zhang^a, Yuping Zhang^a, Bin Xu^a, Leijing Liu^{a*}, Gang Chen^b, Chan Im^c and Wenjing Tian^{a*}

^a State Key Laboratory of Supramolecular Structure and Materials, Jilin University, Changchun 130012, PR China

^b Key Laboratory of Physics and Technology for Advanced Batteries, Ministry of Education, College of Physics, Jilin University, Changchun 130012, PR China.

^c Department of Chemistry, Konkuk University, Seoul 05029, Korea.

Optical Property

Planck's law of radiation describes the photon density $dn(h\nu)$ in a cavity for photon energy between $h\nu$ and $h\nu+dh\nu$ takes the form²:

$$dn(h\nu) = D(h\nu)f(h\nu) dh\nu$$

Here $D(h\nu)$ is the density of the states which the photons can occupy, and $f(h\nu)$ is the distribution function which determines the distribution of photons over the states as a function of the energy $h\nu$. Photons are particles of light, and the fact that we can describe light as an electromagnetic wave is in no way contradictory. Thus the photon density is comprised of optical electric-field distribution and density. According to the studies about the optical modelling of organic solar cell device reported by Persson et al^{3,4} and Hoppe et al⁵, The multilayer of device is treated as a one-dimensional system in which the optical electric field amplitude is calculated as a function of position inside the device starting from the glass-ITO interface. The square of the field strength of electromagnetic wave describes the location of the photons. And the total electric field at an arbitrary position inside layer can be calculated coherently. The optical electric field generates excitons within the solar cell active region. thus the light harvesting which contributes to exciton generation is closely related to the optical electric field distribution and intensity within the PeSC devices⁶.

Table S1 The obtained Rs and Rsh for PeSCs with HTLs of P3HT and Spiro.

P3HT HTL	1	2	3	4	5	6	7	8	9	10
Rs ($\Omega \cdot \text{cm}^2$)	10.12	10.05	9.56	9.21	9.63	10.25	10.06	10.29	10.35	9.19
Rsh ($\Omega \cdot \text{cm}^2$)	808.82	801.21	811.15	815.69	812.74	820.63	805.62	798.56	799.61	815.28
	11	12	13	14	15	16	17	18	19	20
Rs ($\Omega \cdot \text{cm}^2$)	9.89	9.86	10.11	10.03	9.96	10.23	10.42	10.35	9.94	10.26
Rsh ($\Omega \cdot \text{cm}^2$)	802.69	814.56	795.23	798.95	797.58	815.24	805.91	800.05	821.61	816.25
SPIro HTL	1	2	3	4	5	6	7	8	9	10
Rs ($\Omega \cdot \text{cm}^2$)	4.21	4.11	3.96	3.85	4.05	4.25	3.89	4.38	4.61	3.81
Rsh ($\Omega \cdot \text{cm}^2$)	1635.21	1630.12	1632.51	1642.62	1640.25	1620.36	1652.56	1642.31	1638.95	1629.01
	11	12	13	14	15	16	17	18	19	20
Rs ($\Omega \cdot \text{cm}^2$)	4.91	4.46	4.42	3.75	3.82	4.12	4.01	4.26	4.01	4.75
Rsh ($\Omega \cdot \text{cm}^2$)	1625.63	1634.52	1642.35	1635.96	1645.63	1648.25	1654.25	1629.21	1624.52	1638.57

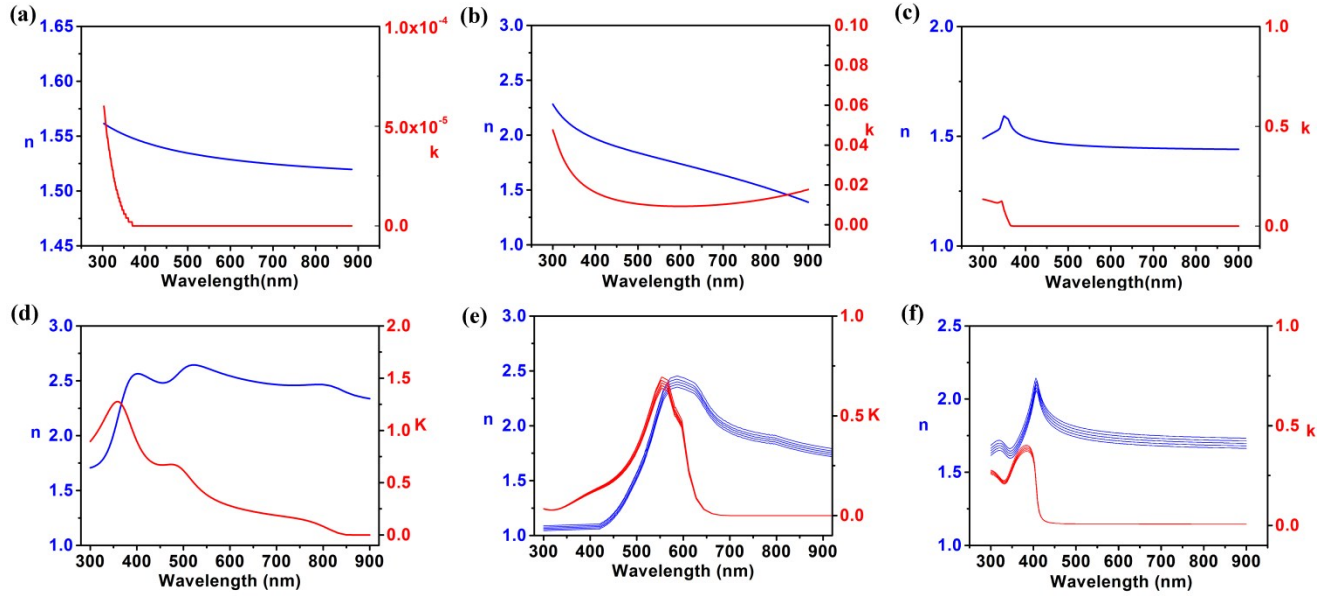


Fig. S1 Complex refractive index spectra for the layers of (a) glass, (b) ITO, (c) ZnO, (d) FAPbI₃, (e) P3HT, (f) Spiro.

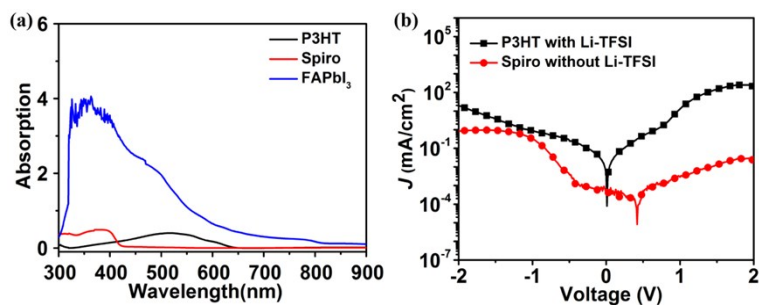


Fig. S2 (a) the absorption of P3HT, Spiro and FAPbI₃ films; (b) J–V characteristics under dark condition measured for the PeSCs with HTLs of P3HT doped with Li-TFSI and pristine Spiro films.

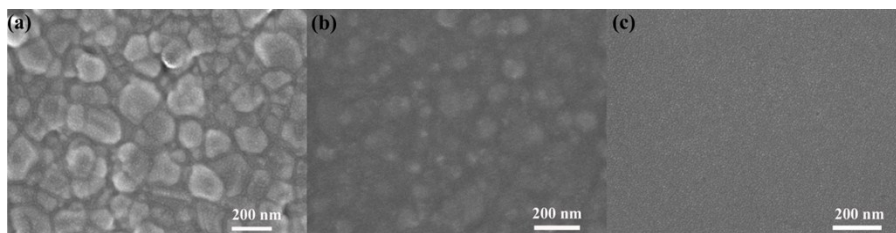


Fig. S3 Top-view SEM images of perovskite film, P3HT and Spiro films covered on the perovskite layers.

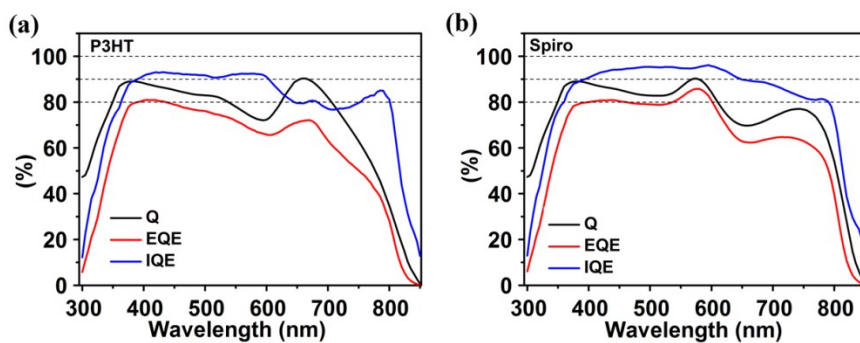


Fig. S4 The Q, EQE and IQE spectra for PeSCs with HTL of P3HT (a) and Spiro (b).

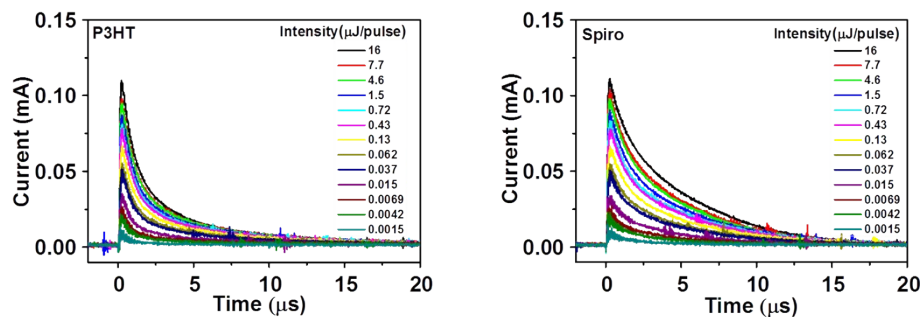


Fig. S5 The intensity-dependent transient photocurrents for PeSCs with HTL of P3HT and Spiro.

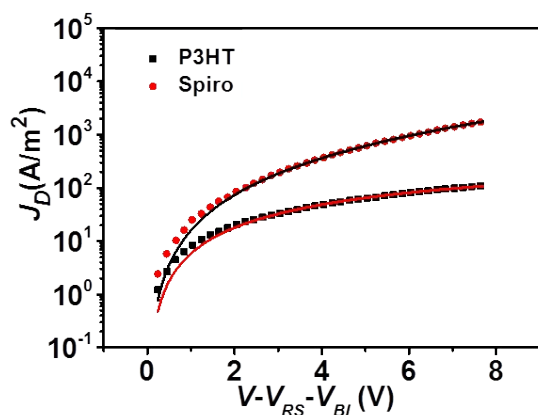


Fig. S6 Experimental dark-current of the P3HT and Spiro film measured at room temperature in the hole-only device, configuration was ITO/P3HT (50nm) or Spiro (150 nm) /Au. The solid lines represent the fit using a model of single-carrier SCL current⁶.

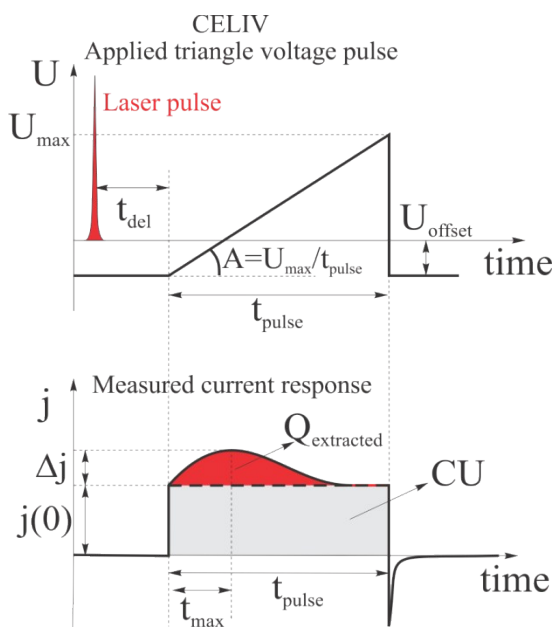


Fig. S7 Schematic illustration of the photo-CELIV method.

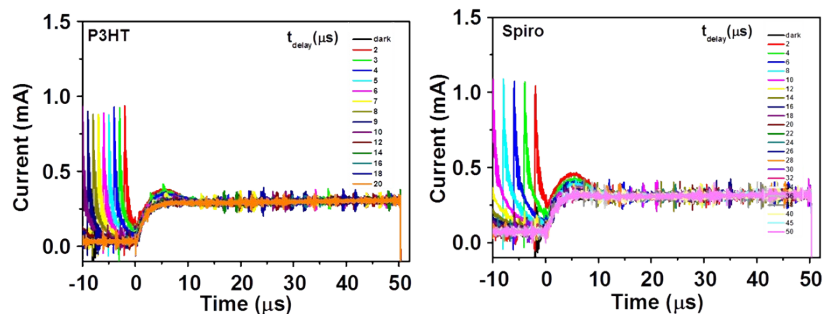


Fig. S8 The photo-CELIV transients at varying delay times for PeSCs with HTL of P3HT and Spiro.

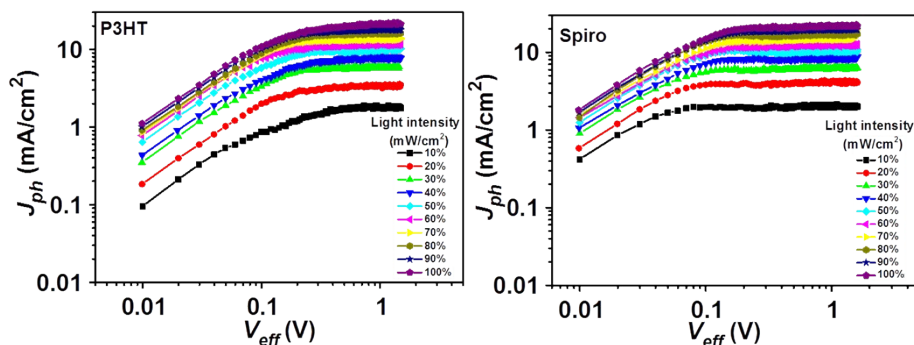


Fig. S9 J_{ph} – V_{eff} characteristics various light intensity for PeSCs with HTL of P3HT and Spiro.

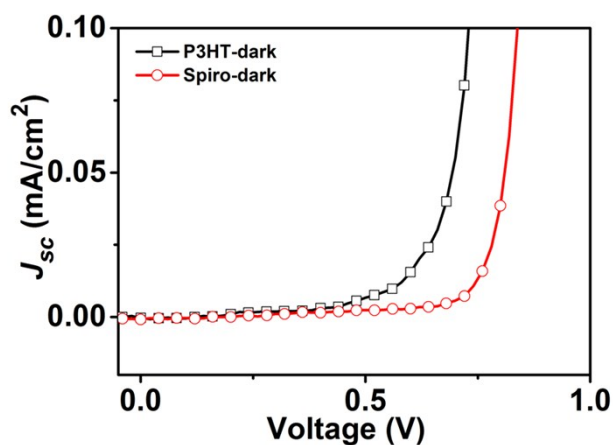


Fig. S10 the J–V characteristics under dark condition for the PeSCs with HTL of P3HT and Spiro.

Table S2 the parameter after fitting , where $n(0)$ is the initial charge carrier density, τ is the recombination lifetime, γ ($0 < \gamma \leq 1$) is the dispersion parameter.

	n_0 (cm ⁻¹)	τ (μs)	γ
P3HT	7.94×10^{15}	7.82	1.0
Spiro	9.97×10^{16}	9.18	1.0

Reference

- 1 Peter Würfel, *Physics of Solar cells – from Principles to New Concepts*, Wiley-VCH, 2005, ISBN: 3-527-40428-7.
- 2 N.K. Persson, O. Inganäs, Organic tandem solar cells—modelling and predictions, *Solar Energy Materials & Solar Cells*, 2006, 90, 3491–3507.
- 3 N.K. Persson, M. Schubert, O. Inganäs, Optical modelling of a layered photovoltaic device with a polyfluorene derivative/fullerene as the active layer, *Solar Energy Materials & Solar Cells*, 2004, 83, 169–186.
- 4 H. Hoppe, N. Arnold, D. Meissner, N.S. Sariciftci, Modeling of optical absorption in conjugated polymeryfullerene bulk-heterojunction plastic solar cells, *Thin Solid Films*, 2004, 451–452, 589–592.
- 5 J. M. Ball, S. D. Stranks, M. T. Hörantner, S. Hüttner, W. Zhang, E. J. W. Crossland, I. Ramirez, M. Riede, M. B. Johnston, R. H. Friend and H. J. Snaith, *Energy Environ. Sci.*, 2015, 8, 602–609.
- 6 V. D. Mihailetschi, H. Xie, B. d. Boer, L. J. Koster, P. W. M. Blom, Charge Transport and Photocurrent Generation in Poly(3-hexylthiophene):Methanofullerene Bulk-Heterojunction Solar Cells, *Adv. Funct. Mater.*, 2006, 16, 699-708.