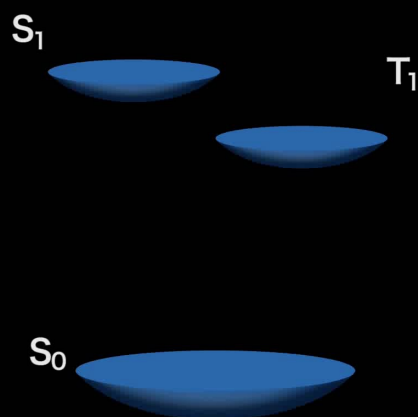




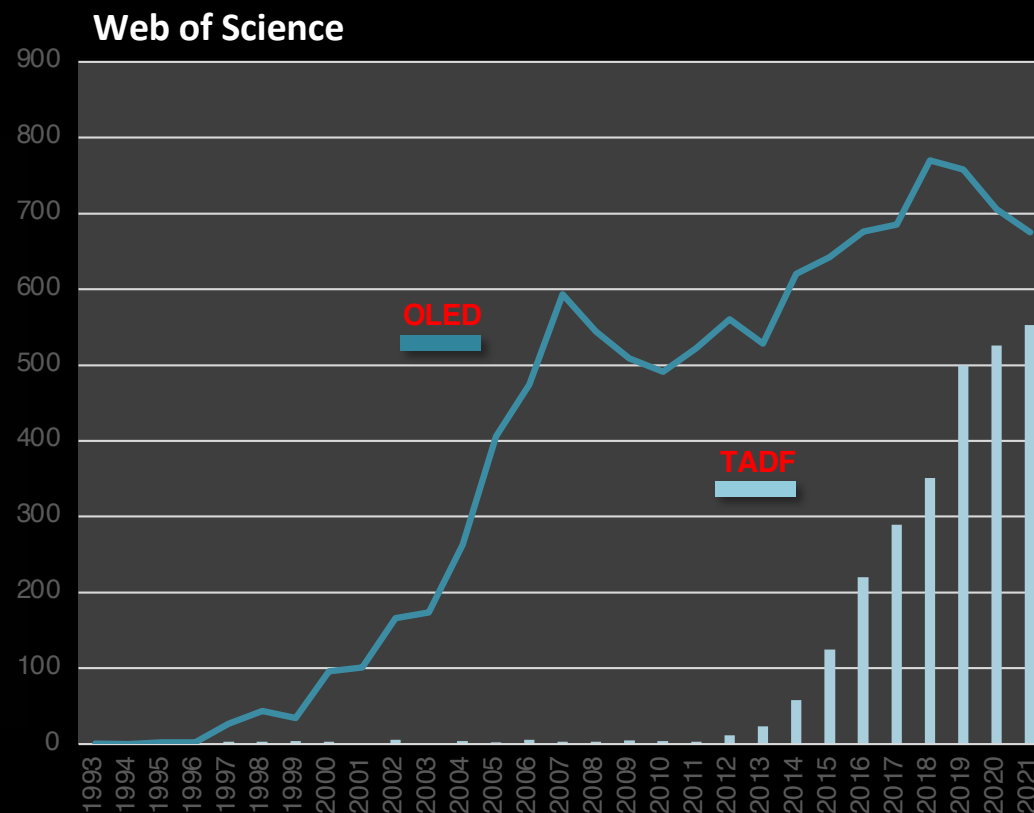
Blue Hyperfluorescence OLEDs aimed for high efficiency and stability

Center for Organic Photonics and Electronics Research (OPERA), Kyushu University

Chin-Yiu Chan, Yi-Ting Lee, Youichi Tsuchiya, Masaki Tanaka, Hajime Nakanotani and Chihaya Adachi

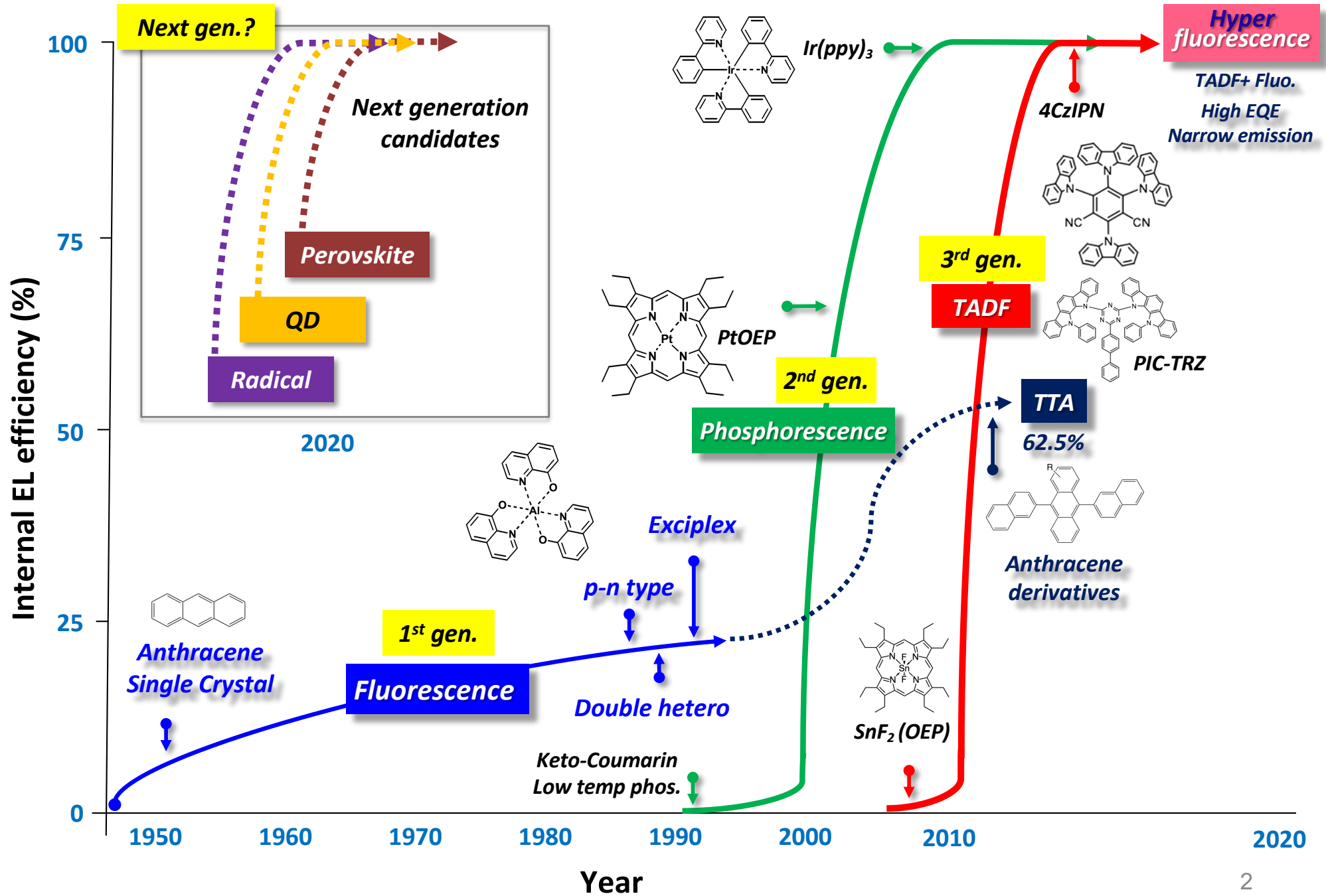


Conventional fluorescence and phosphorescence processes

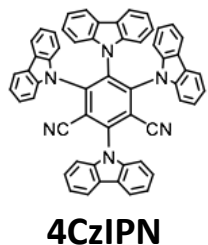


- 1) Importance of short triplet lifetime in OLED emitters
- 2) Blue hyperfluorescence OLEDs with advanced materials combination

Progress of molecular emitters in OLEDs



Acceleration of RISC



$$k_{RISC} = A \exp\left(-\frac{\Delta E_{ST}}{k_B T}\right) \quad A \approx \frac{\langle \phi_i | H_{SO} | \phi_f \rangle}{\Delta E_{ST}} \quad H_{SO} = \xi \cdot S \cdot L$$

A is related with spin orbital coupling (H_{SOC})

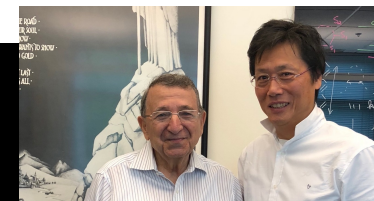
ξ : spin-orbit coupling constant (Heavy atom effect)

L : orbital angular momentum

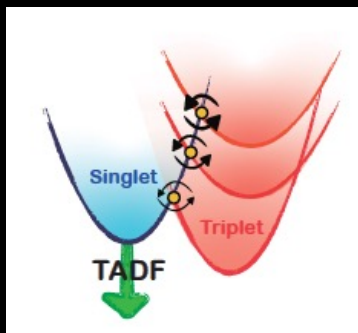
S : spin angular momentum

El-Sayed rule: Formulated by Prof. Mostafa A. El-Sayed in 1960s

Chemical Reviews 66 (2): 199–241.



Mostafa



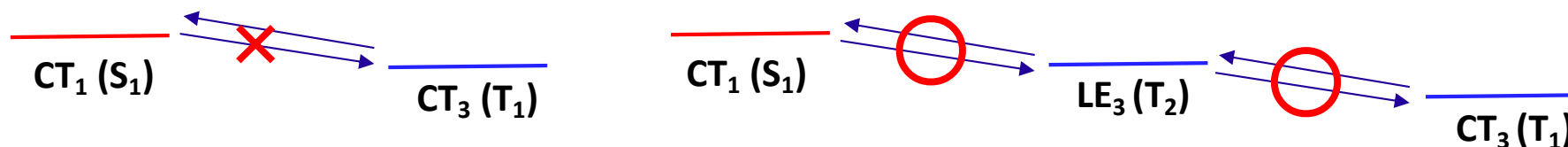
$${}^1\text{CT} \overset{\times}{\longleftrightarrow} {}^3\text{CT} \quad {}^1\text{CT} \overset{\circ}{\longleftrightarrow} {}^3\text{LE}$$

$$\langle \psi_{1CT} | H_{SOC} | \psi_{3CT} \rangle = 0 \quad \langle \psi_{1CT} | H_{SOC} | \psi_{3LE} \rangle \neq 0$$

Second order vibronic coupling between LE and CT states

$$k_{RISC} = \frac{2}{\hbar} \left| \frac{\langle \psi_{1CT} | H_{SOC} | \psi_{3LE} \rangle \langle \psi_{3LE} | H_{vib} | \psi_{3CT} \rangle}{E_{3CT} - E_{3LE}} \right|^2 \delta(E_{1CT} - E_{3LE})$$

M. K. Etherington et al., *Nat. Commun.*, 7, 13680 (2016)



Quantum Chemical Cal.: Excited-state engineering for efficient TADF



Prof. Xiankai Chen
(now, City U of HK)

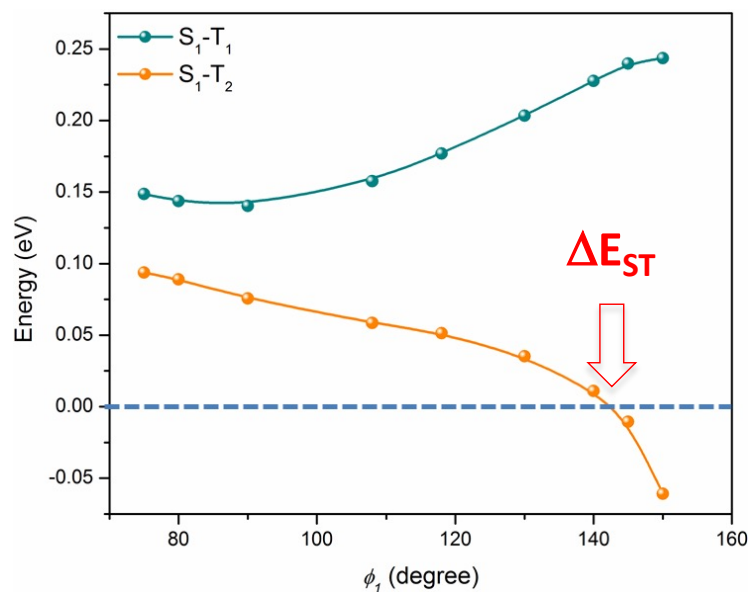
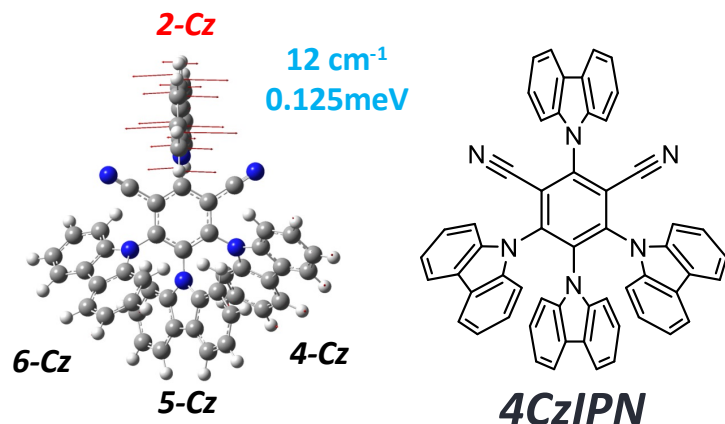
Article | Published: 02 September 2019

Critical role of intermediate electronic states for spin-flip processes in charge-transfer-type organic molecules with multiple donors and acceptors

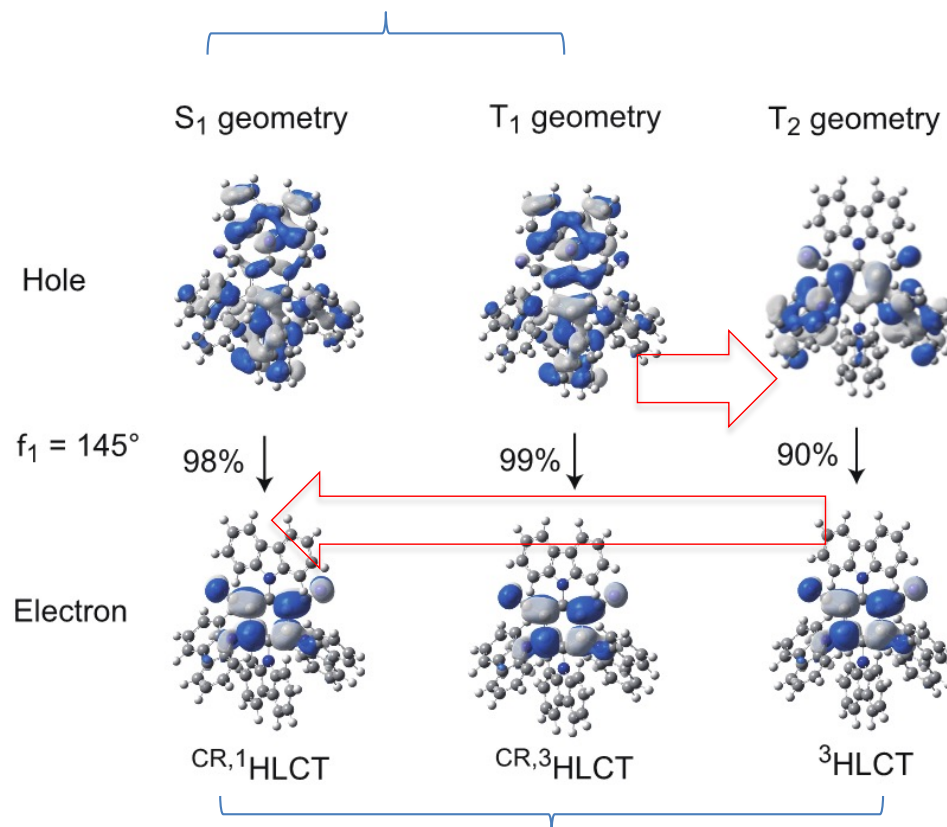
Hiroki Noda, Xian-Kai Chen, Hajime Nakanotani, Takuya Hosokai, Momoka Miyajima, Naoto Notsuka, Yuuki Kashima, Jean-Luc Brédas & Chihaya Adachi

Nature Materials 18, 1084–1090(2019) | Cite this article

9110 Accesses | 35 Citations | 15 Altmetric | Metrics



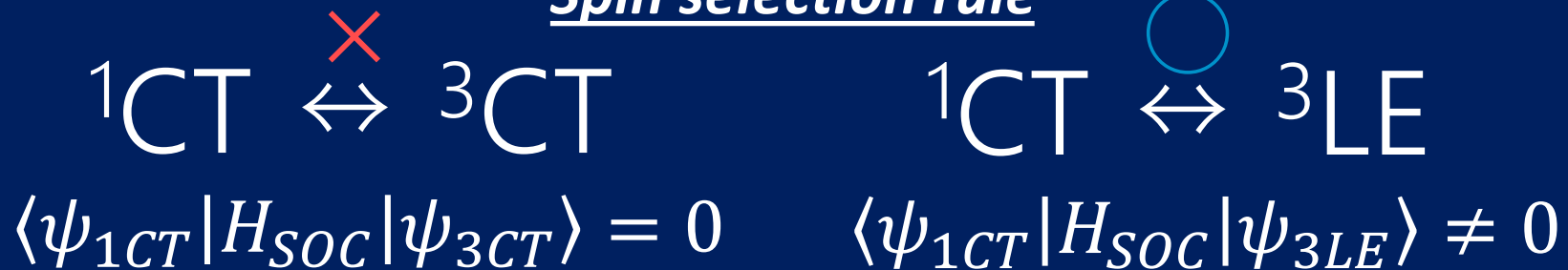
SOC: 0.06 cm⁻¹ S_1-T_1 (CR,¹HLCT-³HLCT)



SOC: 0.51 cm⁻¹

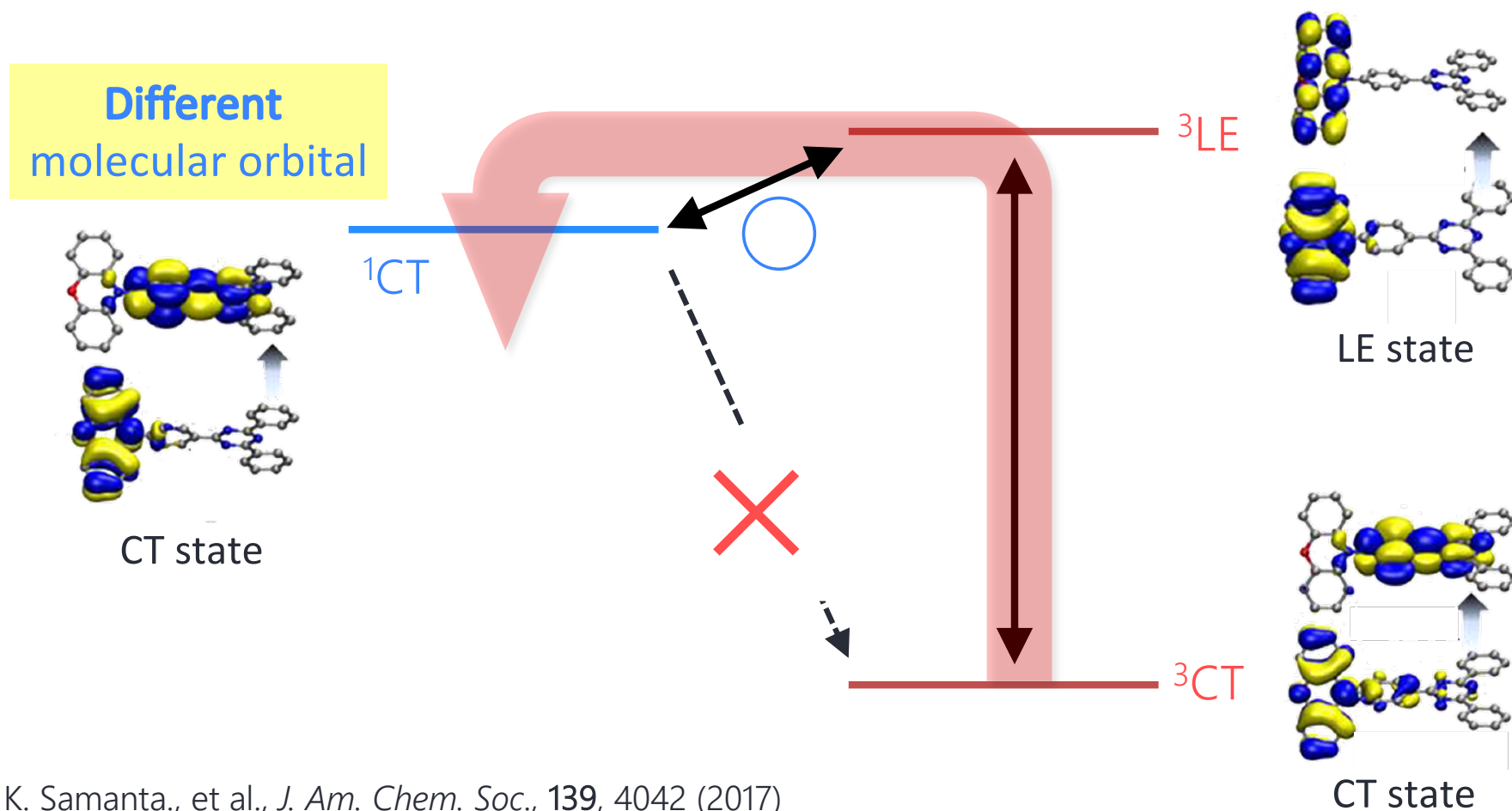
S_1-T_2 (CR,¹HLCT-³HLCT)

Spin selection rule



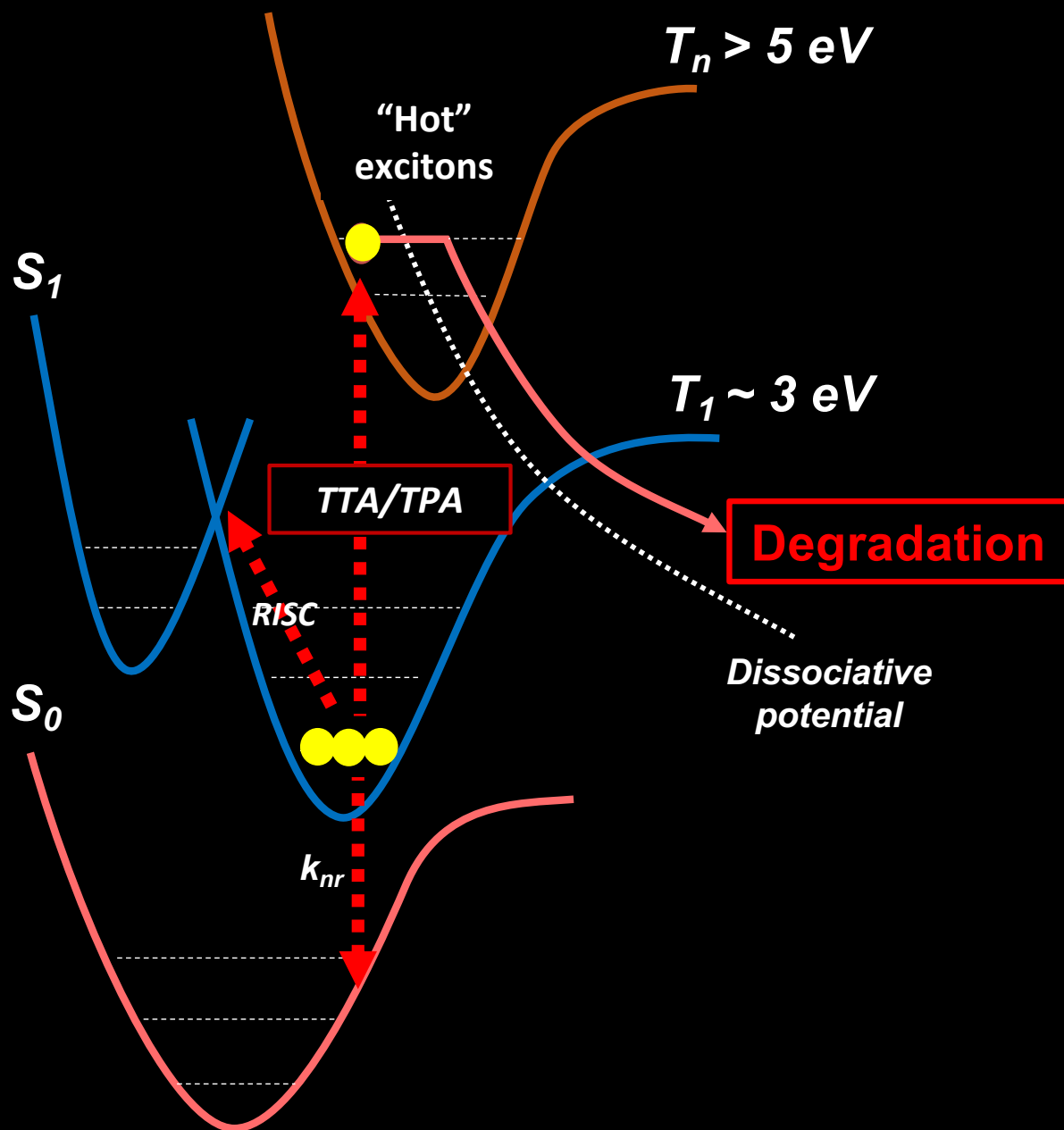
M. A. El-Sayed, et al., *J. Chem. Phys.*, **36**, 573 (1962).

B. T. Lim, et al., *Chem. Phys. Lett.*, **79**, 22 (1981).

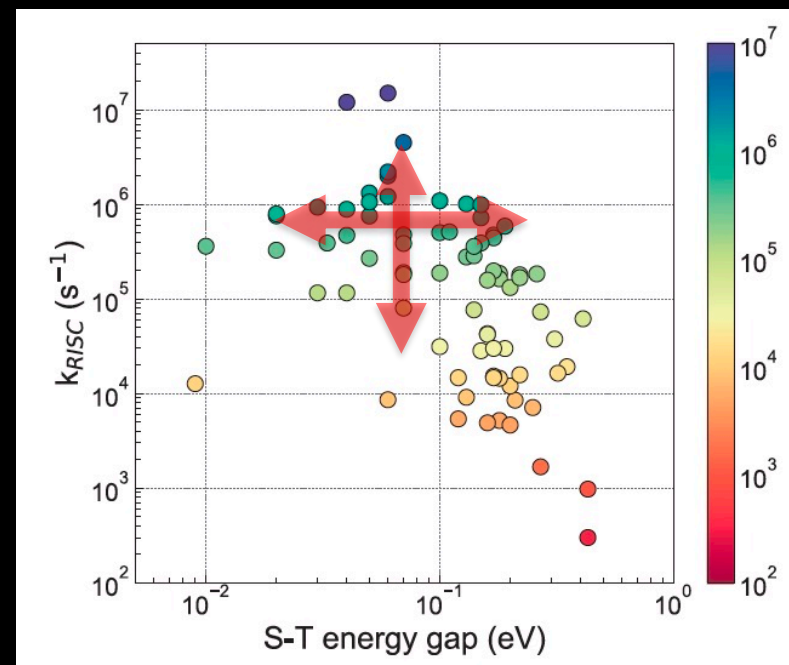


P. K. Samanta, et al., *J. Am. Chem. Soc.*, **139**, 4042 (2017)

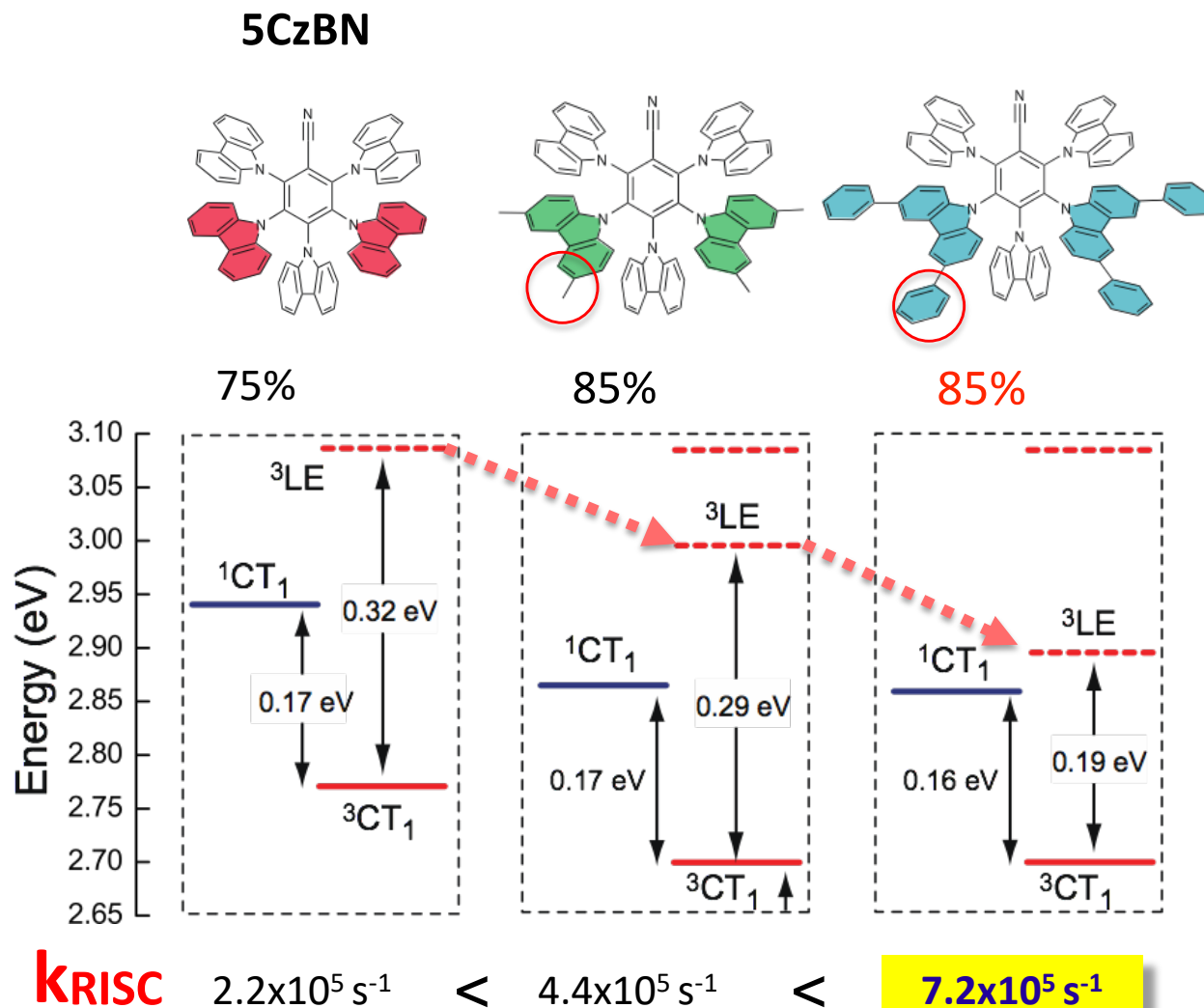
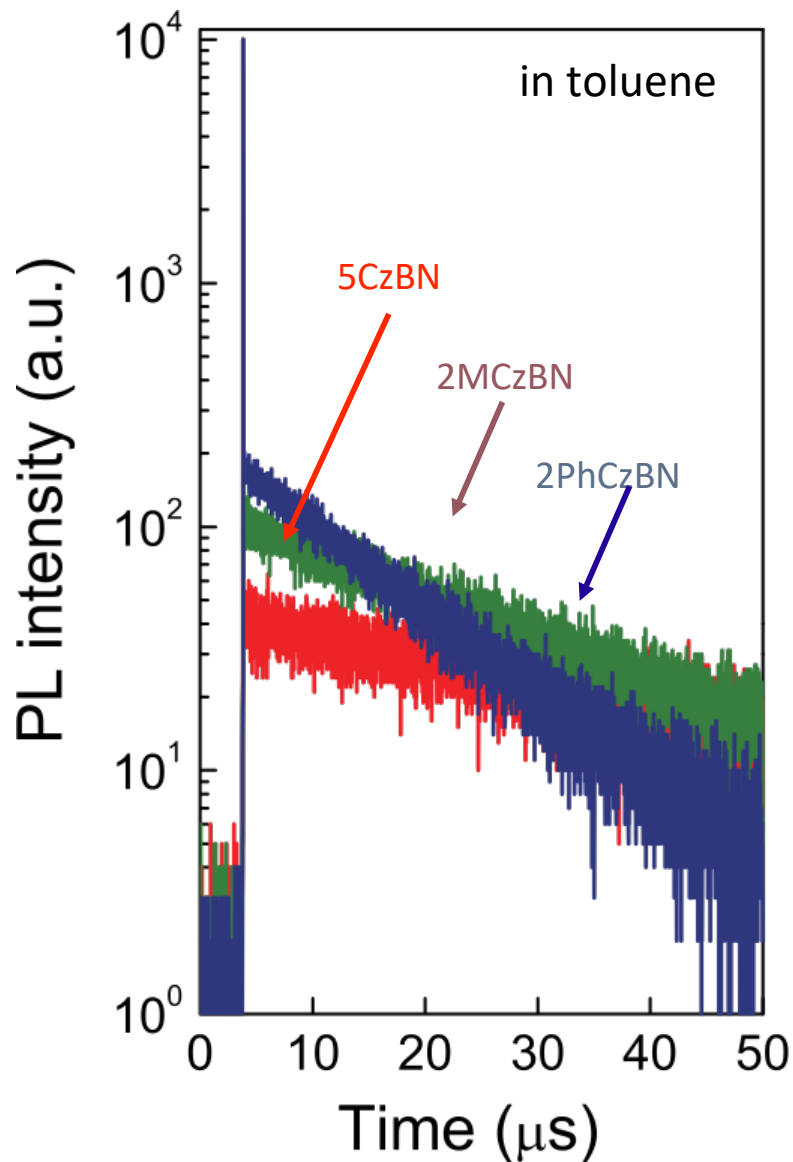
OLED device lifetime - Degradation via triplet states -



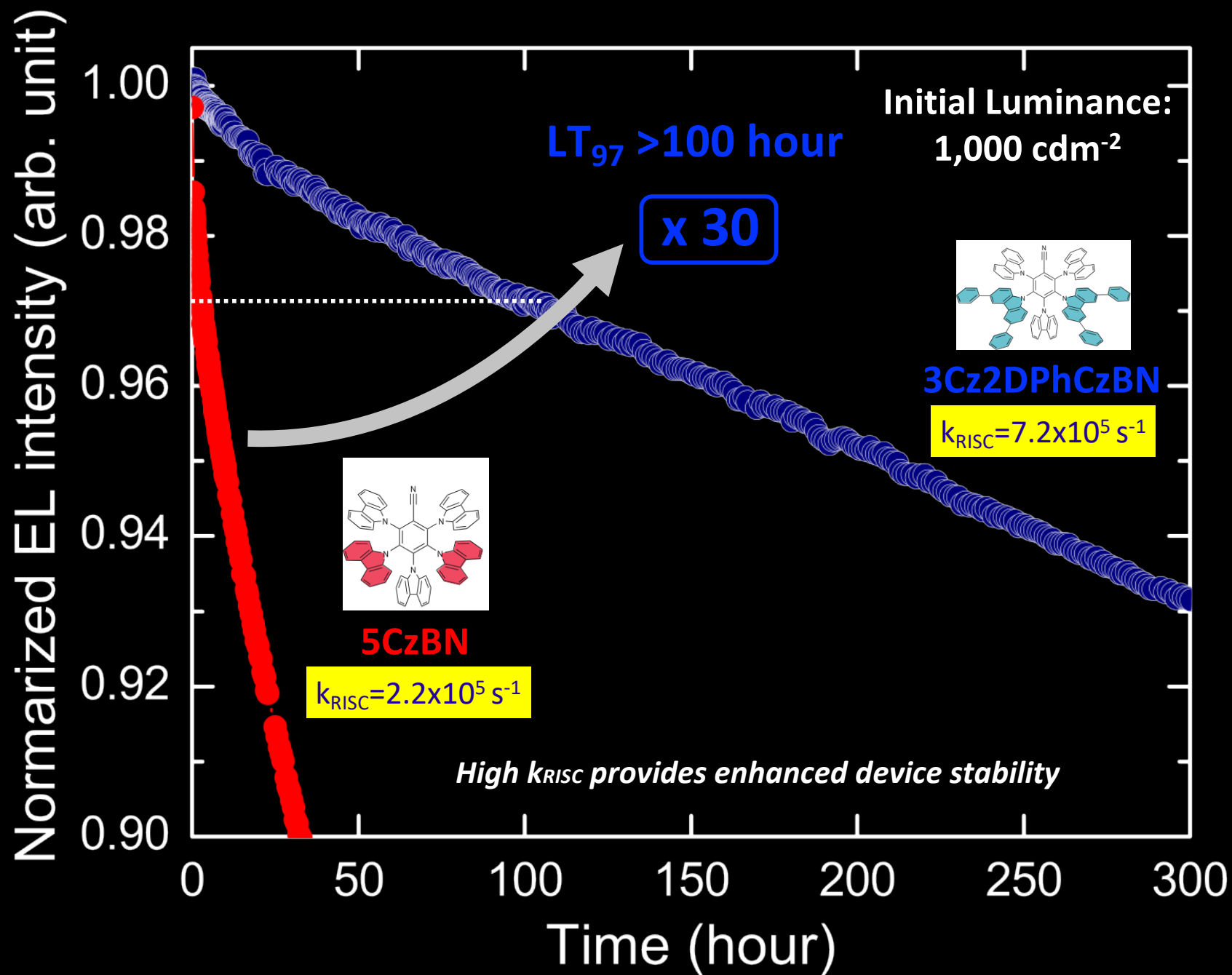
- ❖ Long-live triplet exciton
 - ❖ μs to ms order
 - ❖ Exciton annihilation
- ❖ High triplet energy
 - ❖ $T_1 \sim 3 \text{ eV}$ (for blue)
 - ❖ $T_n > 5 \text{ eV}$ (for blue)
- ❖ Chemical bond dissociation
 - ❖ C-N: $\sim 3.5 \text{ eV}$



Excited-state engineering for efficient TADF



Prolonged OLED device lifetime with

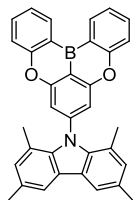


Recent advanced TADF emitters

$$k_{ISC} > k_{RISC} > k_r$$

TMCz-BO

$$k_r > k_{ISC} > k_{RISC}$$



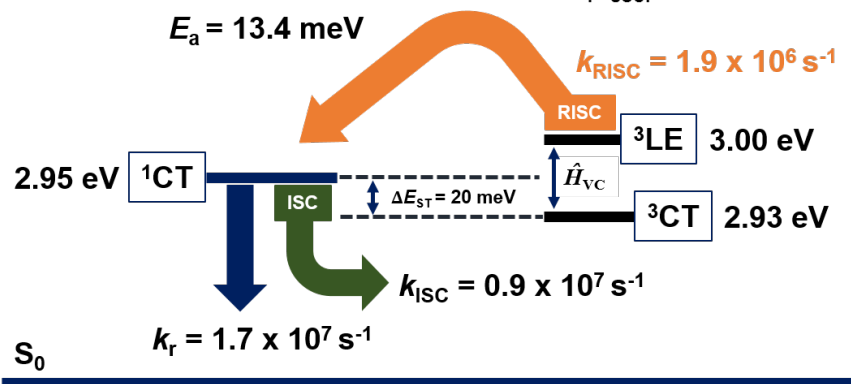
JU Kim et al., Nature Comm. 11, 1765 (2020)

(a) TMCz-BO

$\tau_d = 750$ ns

$$\langle {}^1CT | \hat{H}_{soc} | {}^3LE \rangle = 0.124 \text{ cm}^{-1}$$

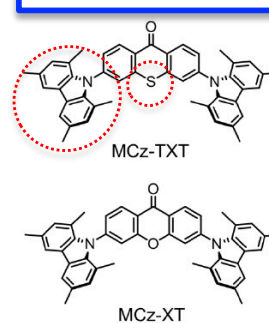
$$\langle {}^1CT | \hat{H}_{soc} | {}^3CT \rangle = 0.001 \text{ cm}^{-1}$$



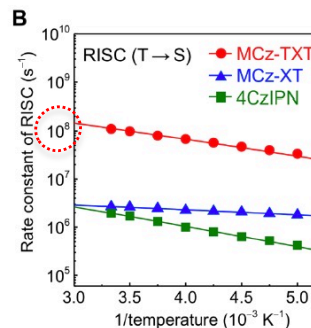
MCz-TXT

N. Aizawa, et al., *Sci. Adv.* 7, eabe5769 (2021)

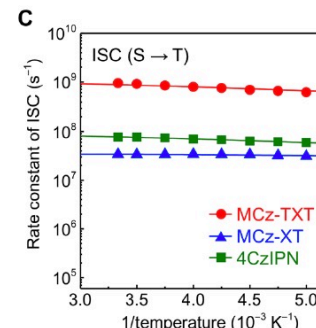
A



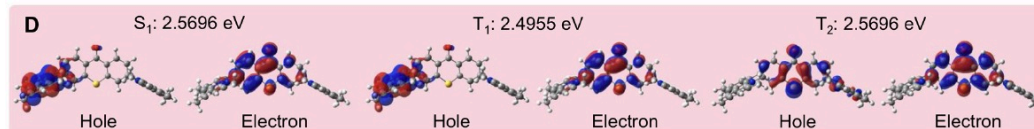
B



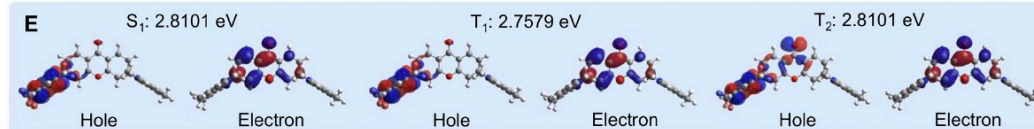
C



D

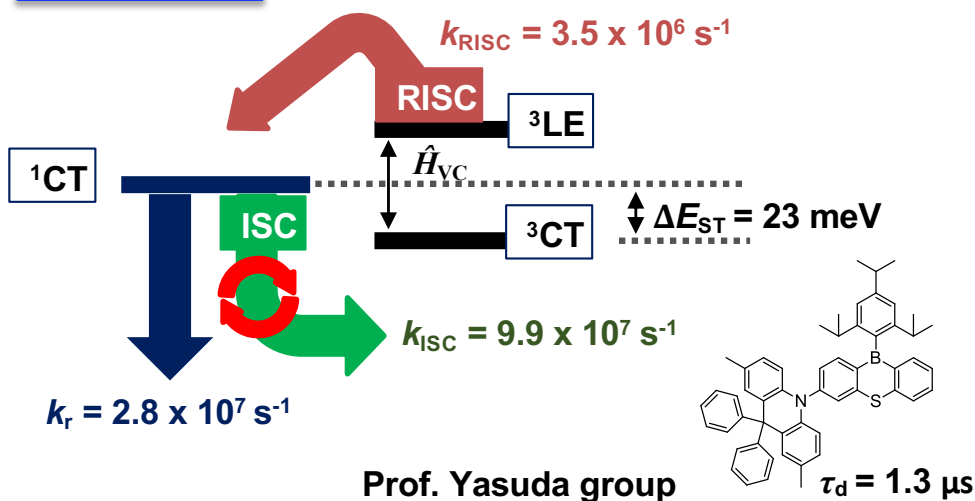


E



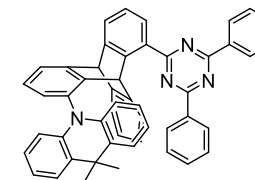
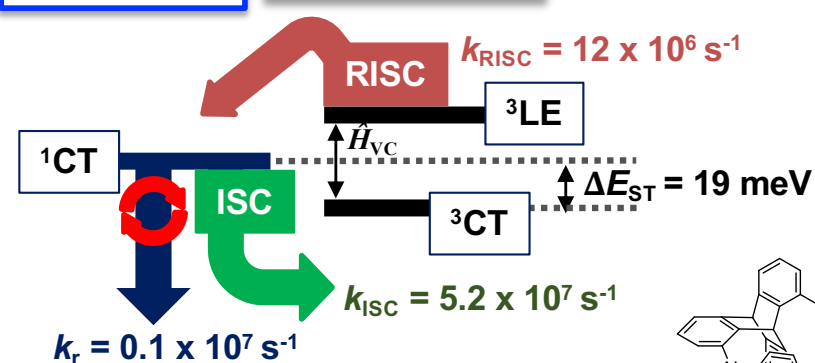
MPAc-BS

$$k_{ISC} > k_r > k_{RISC}$$



TpAT-tFFO

$$k_{ISC} > k_{RISC} > k_r$$

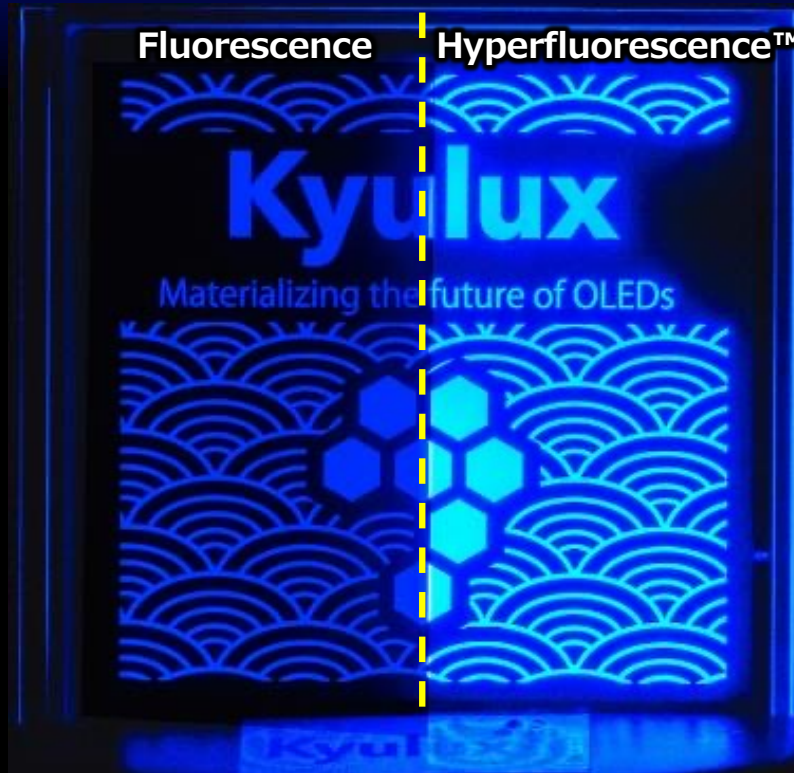
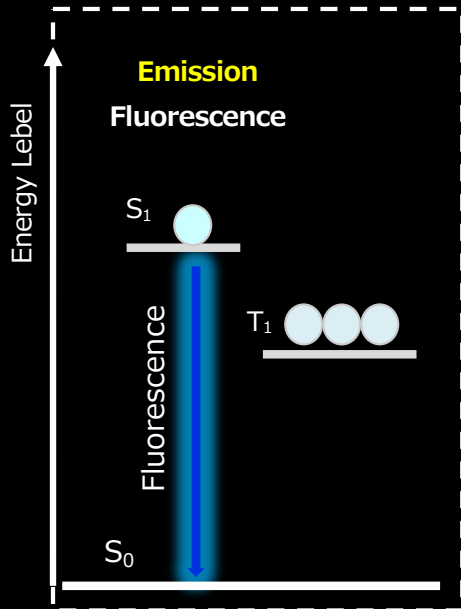


Prof. Kaji group

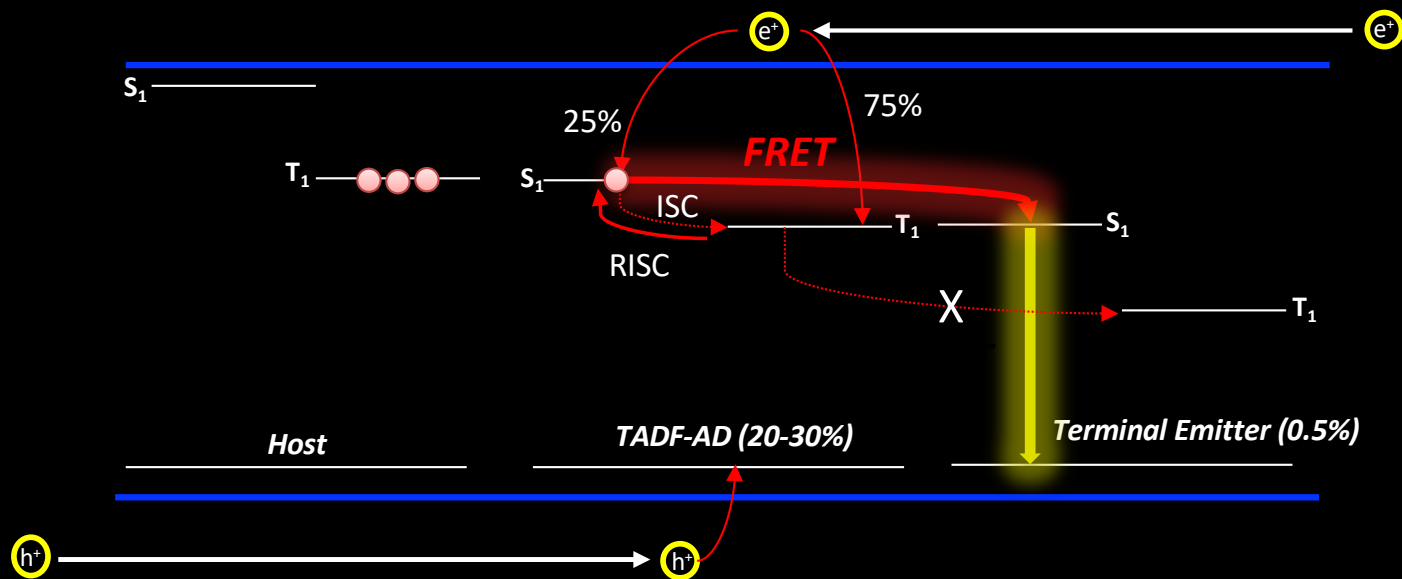
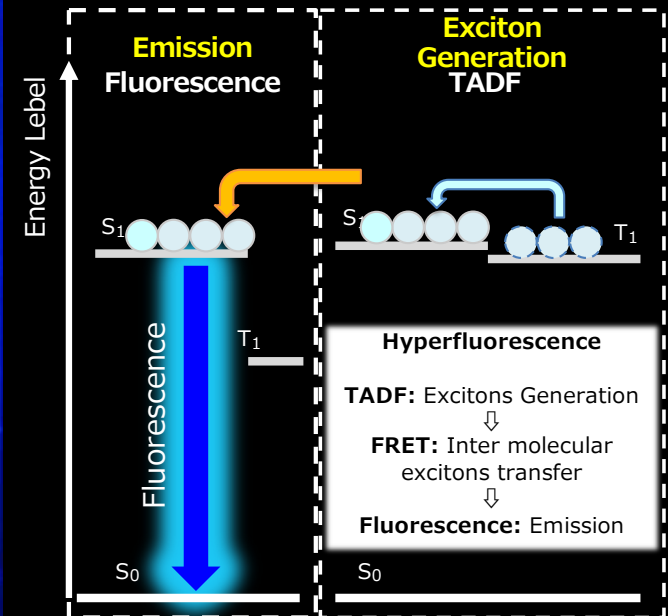
$\tau_d = 4.1$ μs

Advanced Blue Hyperfluorescence OLED

Fluorescence
Low efficiency:
 $IQE_{MAX}=25\%$



Hyperfluorescence
High efficiency: $IQE_{MAX}=100\%$



Hyperfluorescence™: The Optimum Solution for OLED Display

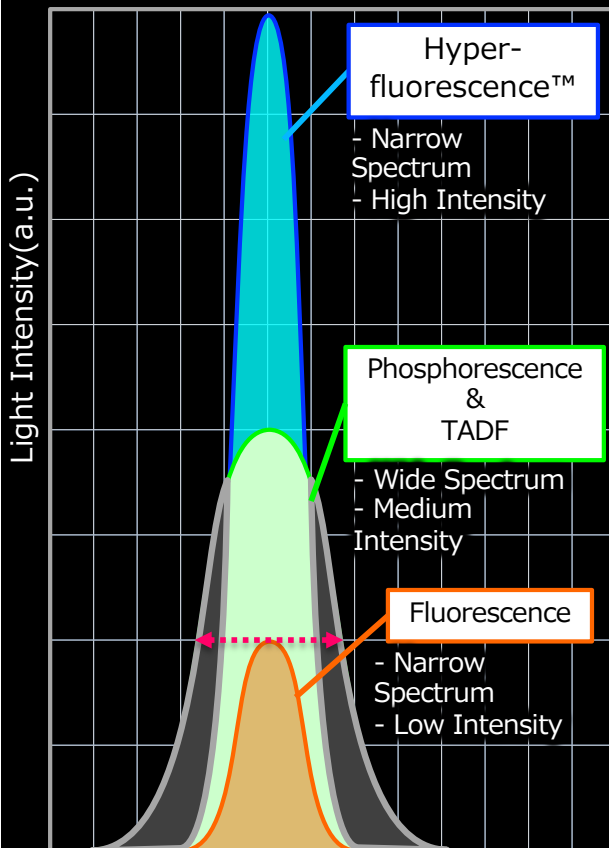
Nat. Commun., 5, 5016, 2014

H. Nakanotani et al.

M. Baldo et al., Nature 403, 750 (2000) Phos. sensitizing fluo.

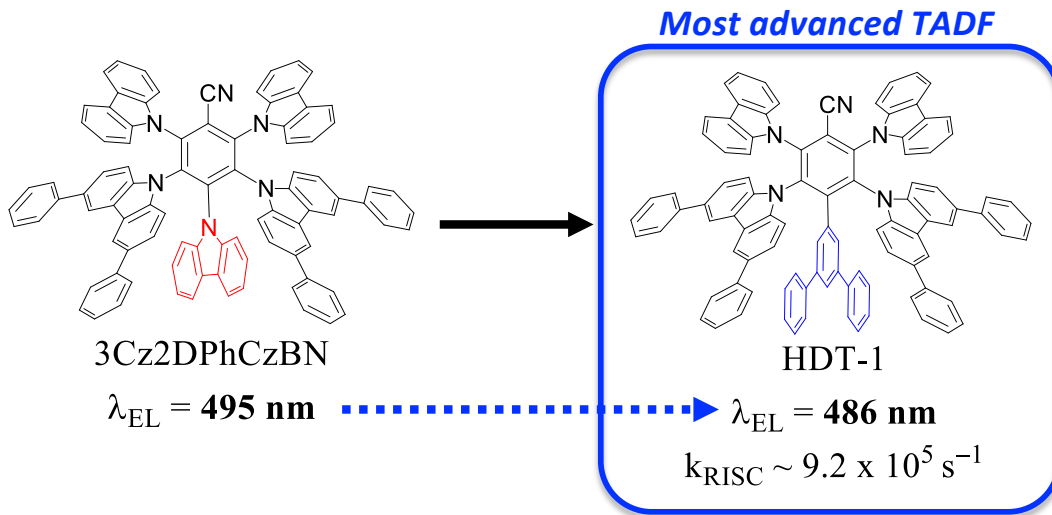
L. Duan et al., Adv. Mat., 26, 5050 (2014) TADF sensitizing fluo.

Spectra Comparison



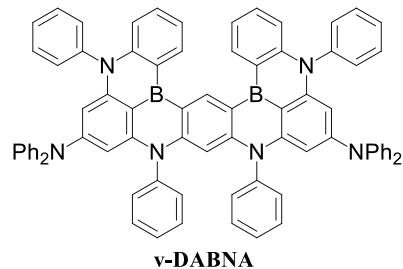
Emission Technology	R	G	B	Efficiency	Cost	Color Purity	
Fluorescence (1Gen)	—	—	○	Low	\$	high	
Phosphorescence (2Gen)	○	○	—	High	\$\$\$	Low	
TADF (3Gen)	○	○	—	High	\$	Low	
Hyperfluorescence™ (4Gen)	○	○	○	High	\$	High	

Advanced Performance of Hyperfluorescence Blue OLED



C-Y. Chan et al.,
Nat. Photonics, 2021
 DOI: <https://doi.org/10.1038/s41566-020-00745-z>

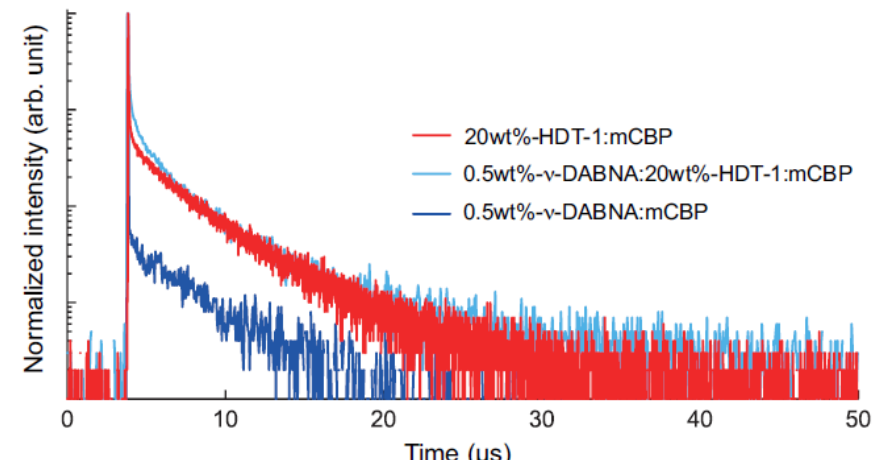
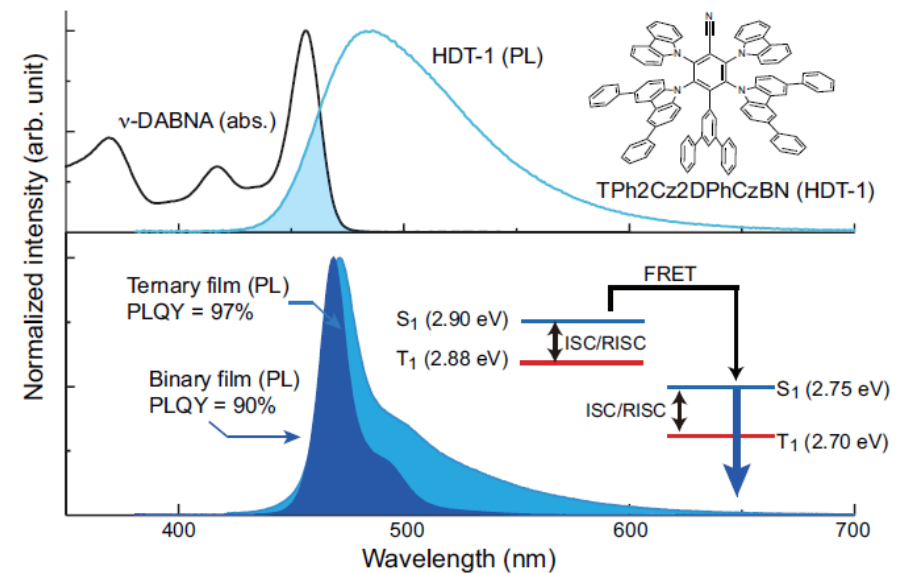
- ◆ Bulky terphenyl substituent prevents molecular aggregation, while maintains a fast k_{RISC} and a high PLQY



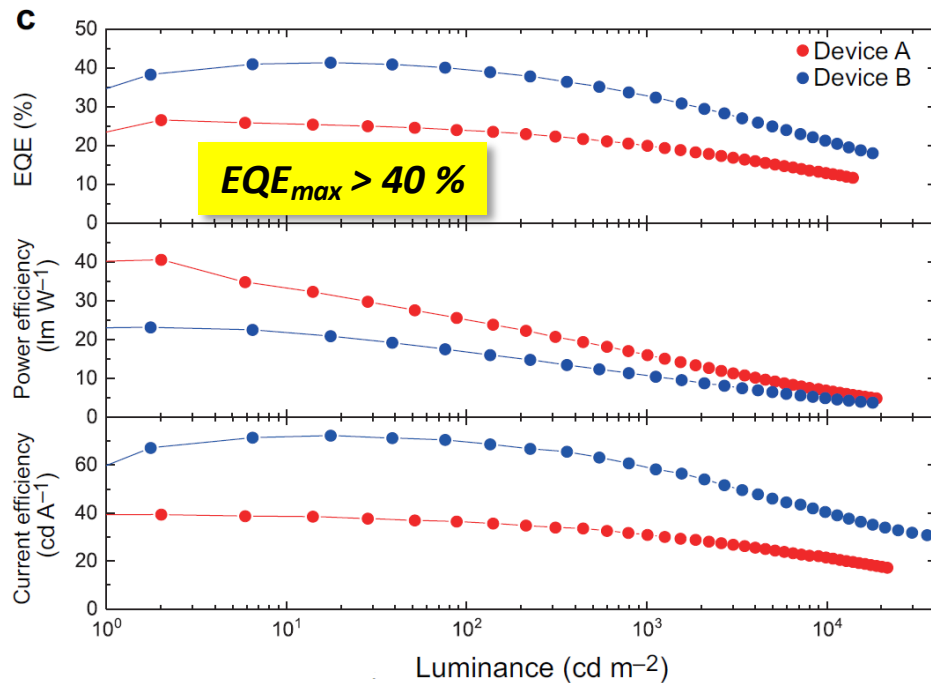
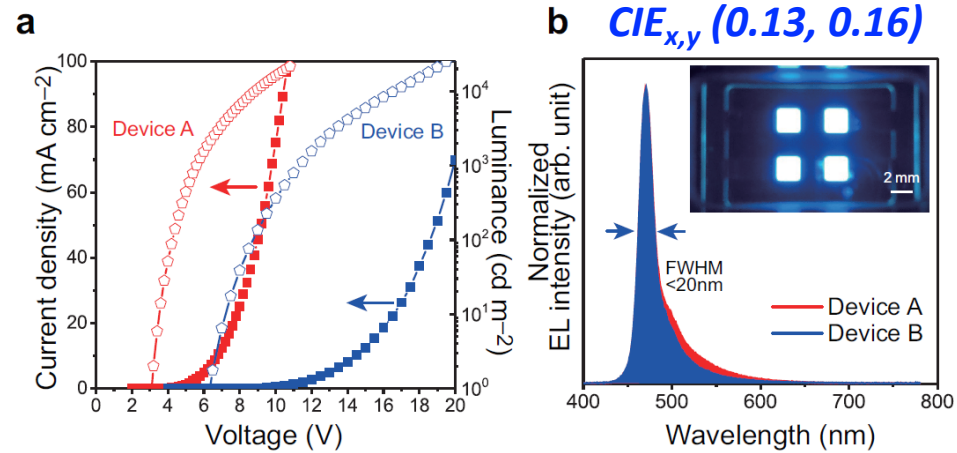
Terminal Emitter

- ◆ Sky-blue emission of HDT-1 ensures singlet-excited state energy transfer to the terminal dopant, hence improving color purity.

T. Hatakeyama and coworkers,
Nat. Photonics 2019, 13, 678.



Hyperfluorescence Blue OLED: Tandem for brighter luminance

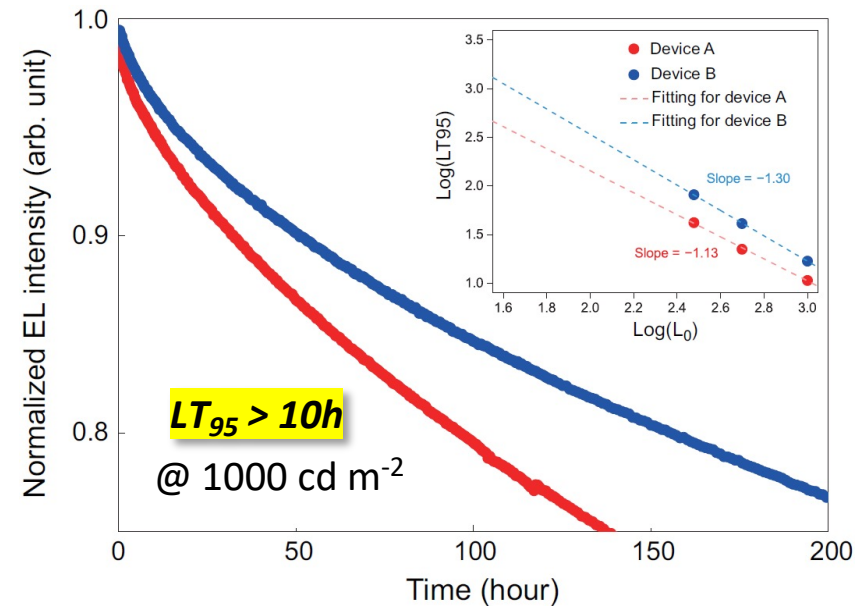


A (Single)

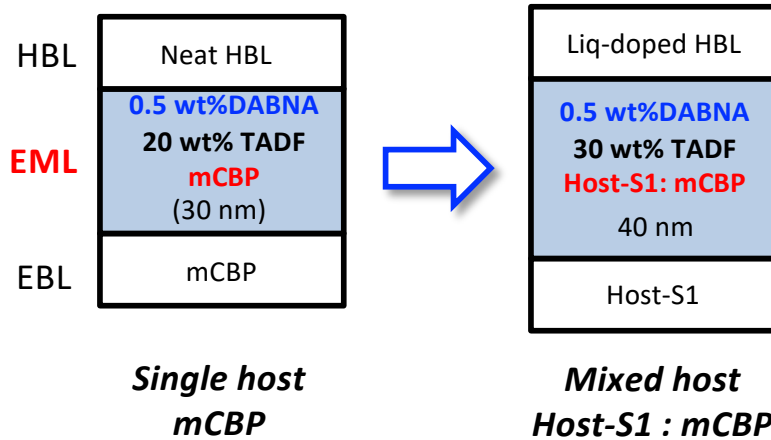
Al (100 nm)
Liq (2 nm)
SF3-TRZ:Liq (25 nm)
SF3-TRZ (10 nm)
EML (30 nm)
mCBP (5 nm)
Tris-Pcz (10 nm)
TAPC (20 nm)
HAT-CN (10 nm)
ITO (50 nm)

B (Tandem)

Al (100 nm)
Liq (2 nm)
SF3-TRZ:Liq (25 nm)
SF3-TRZ (10 nm)
EML (30 nm)
mCBP (5 nm)
Tris-Pcz (10 nm)
TAPC (20 nm)
HAT-CN (10 nm)
Al (1.5 nm)
Liq (2 nm)
SF3-TRZ:Liq (25 nm)
SF3-TRZ (10 nm)
EML (30 nm)
mCBP (5 nm)
Tris-Pcz (10 nm)
TAPC (70 nm)
HAT-CN (10 nm)
ITO (50 nm)

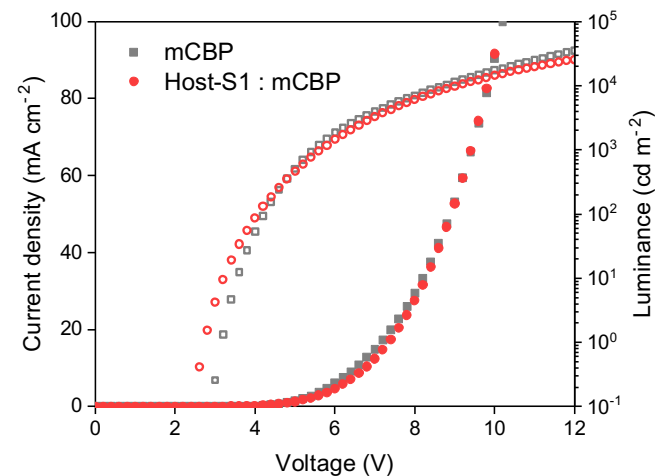
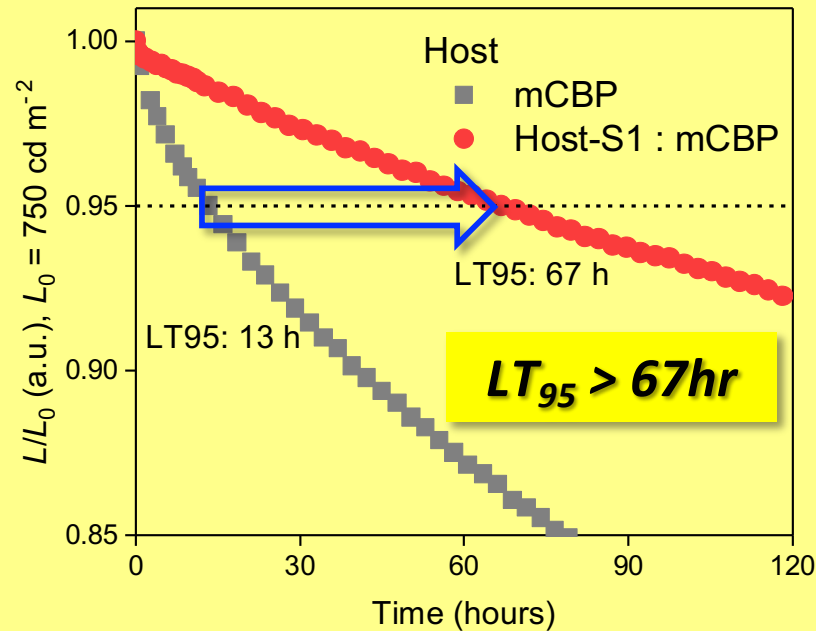
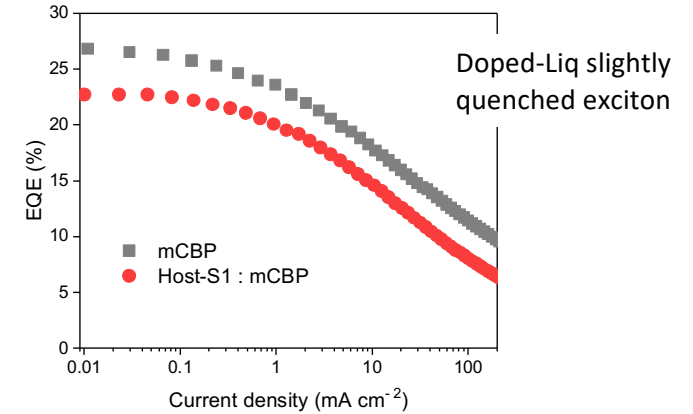
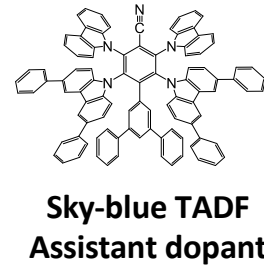
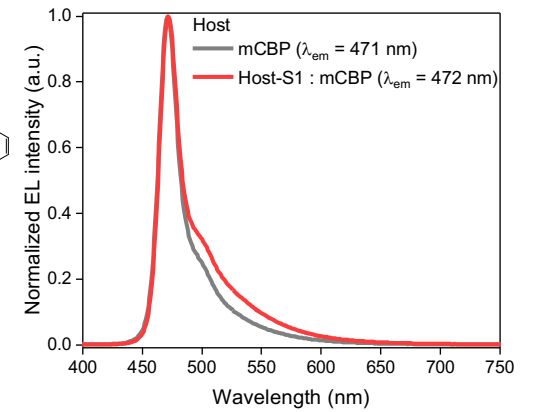
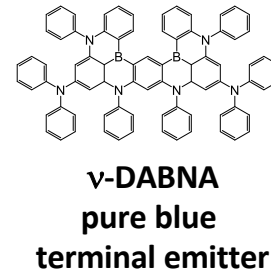
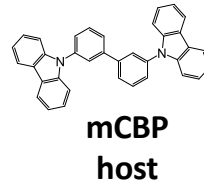


Hyperfluorescence Blue OLED: Lifetime enhancement by *mixed host*

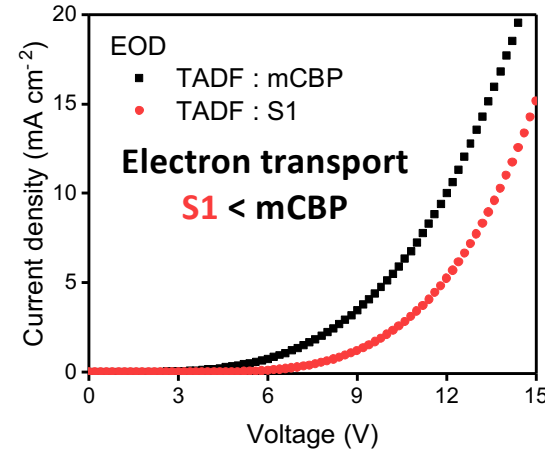
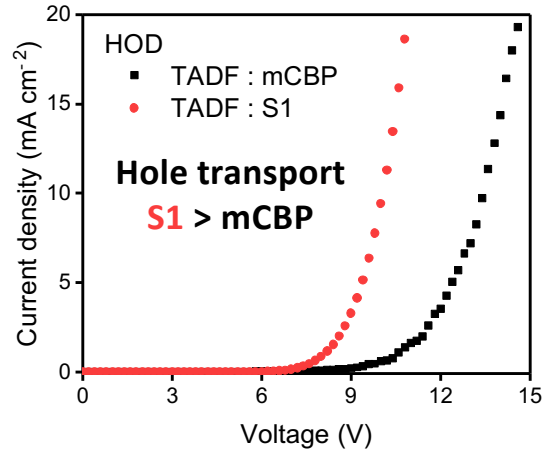


Host-S1

- Cz-based
- Slight shallow HOMO
- High hole-transport

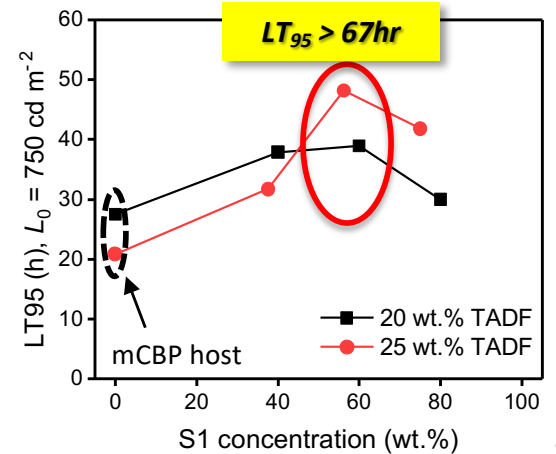
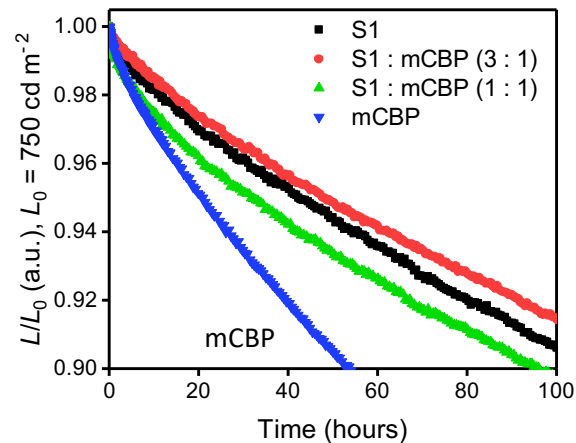
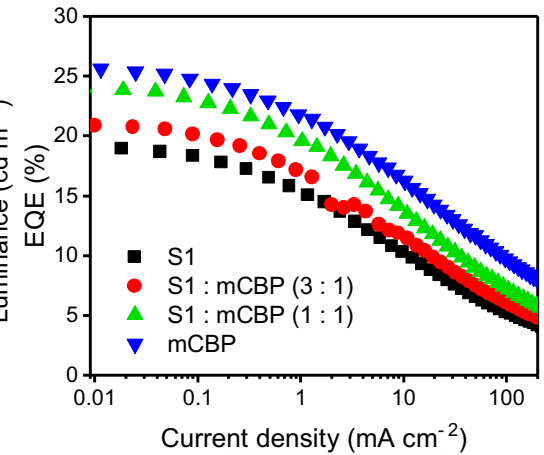
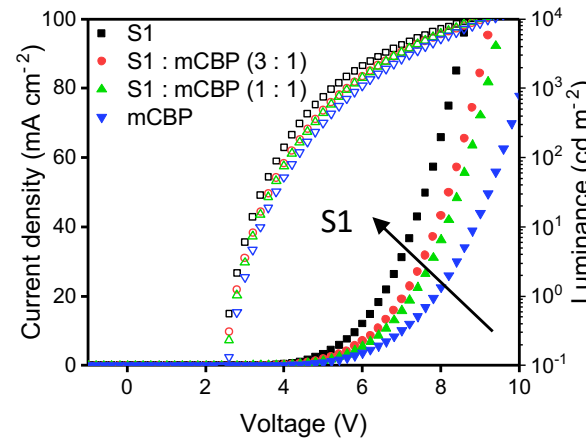
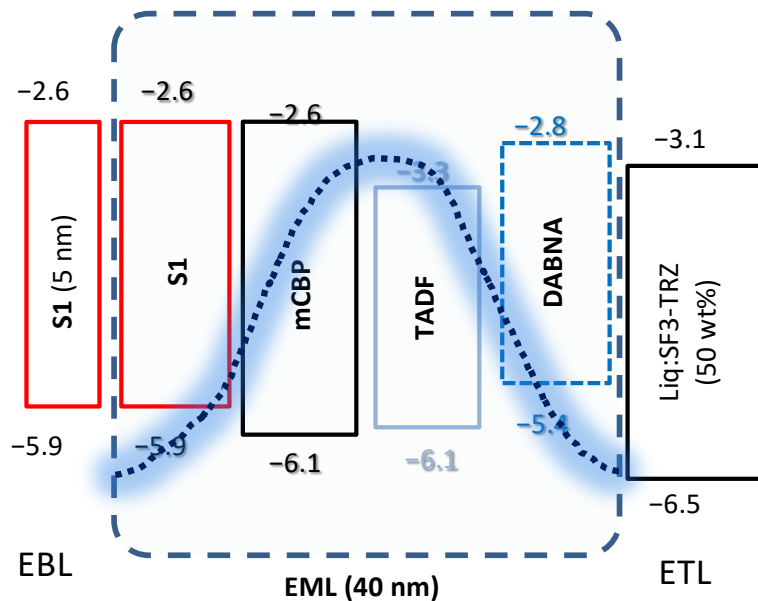


Hyperfluorescence Blue OLED: Lifetime enhancement with mixed host



Host-S1

- Cz backbone
- Shallow HOMO
- $T_1 \sim 2.8$ eV



Latest achievements: Red, Green and Blue



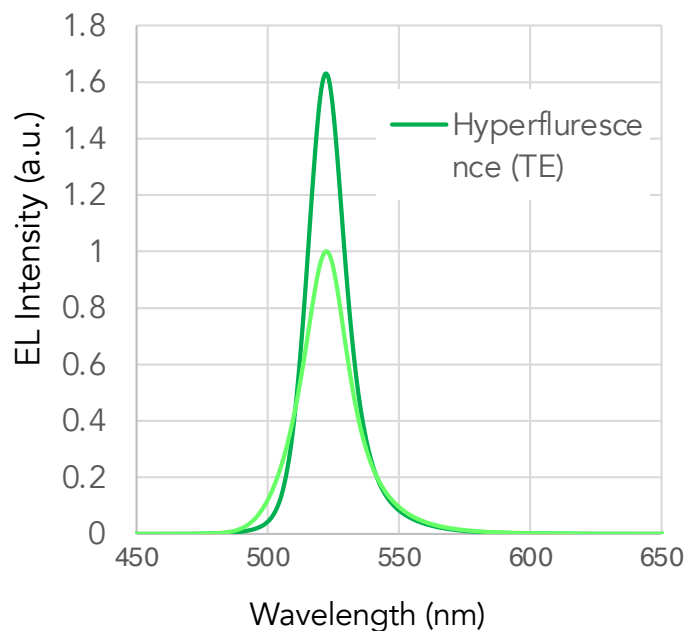
SID Display Week 2021 Symposium

Color		λ_{peak} (nm)	FWHM (nm)	CIE xy	Efficiency (cd/A)	LT95@1000nit (hours)
Red	BE	617	43	0.65, 0.35	32	>37,000
	TE	618	23	0.68, 0.32		
Green	BE	522	34	0.24, 0.70	81	> 20,000
	TE	523	17	0.14, 0.79		
Blue	BE	471	21	0.12, 0.13	43	280
	TE	469	16	0.12, 0.06		

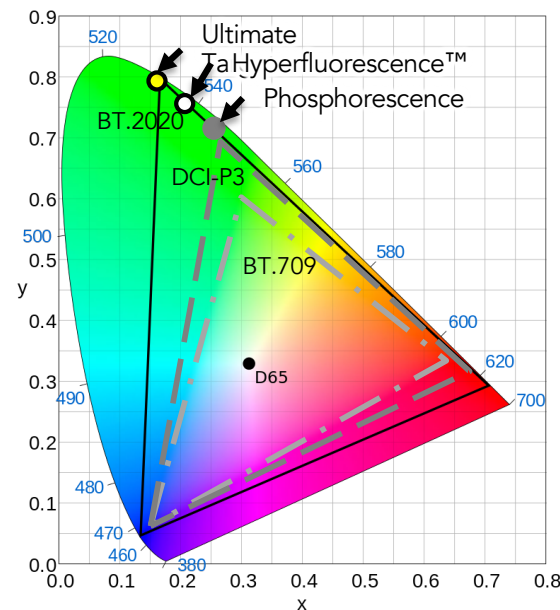


- Top-emitting green Hyperfluorescence™ (HF) device has achieved better chromaticity, higher current efficiency, and longer lifetime than phosphorescence.
- The current target specs in efficiency, lifetime and driving voltage have been achieved.
- Chromaticity of the HF reaches 95% of the ultimate target. The target shall be achieved by using optimized green fluorescence dopants designed by Kyulux before soon.
- HF is able to satisfy the major requirements of the next-generation display. (BT.2020)

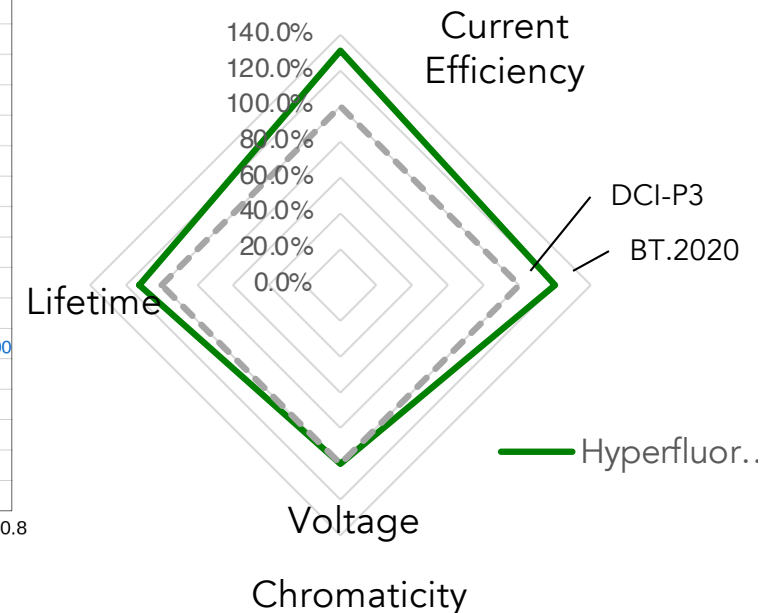
Emission Spectrum of Device



Chromaticity Coordinate



Hyperfluorescence™ Performance



Acknowledgments

TADF works

OPERA members

Prof. H. Nakanotani
Prof. K. Goushi
Prof. T. Matsushima
Prof. R. Kabe (OIST)
Prof. Y. Tsuchiya
Prof. M. Mamada
Prof. T. Yasuda
Prof. T. Nakagawa (YNU)
Prof. S. Hirata (UEC)
Prof. T. Komino (Hyogo)
Prof. J-C. Ribierre
Prof. F. Mathevet (CNRS)
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Dr. Y-T. Lee
Dr. M. Tanaka
Dr. C-Y. Chan
Dr. Yang Gen
Dr. H. Miyazaki
Mr. K. Inada

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Prof. F. Bencheikh
Prof. R. Komatsu
Prof. T. Fujihara
Prof. H. Fujimoto
Prof. H. Miyazaki

All OPERA Students and Staff!

Advisory and Mentors

Prof. K. Tokumaru
Prof. K. Iga
Prof. H. Sasabe
Prof. M. Kotani
Prof. T. Tsutsui
Prof. S. Saito
Prof. Y. Taniguchi
Prof. S. Forrest

TADF Collaboration

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Prof. T. Kawai (NAIST)
Prof. H. Naito (Osaka Pref.)
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Prof. E. Zysman-Coleman (StAndrews)
Prof. J-J. Kim (SNU)
Prof. K-T. Wong (NTU)
Prof. Z. Jiang (Soochow)

Special thanks to All OPERA members!

**Japan Science and Technology Agency JST-ERATO, Regional Innovation Ecosystems MEXT
Kyulux Inc.**

