

XU GROUP  
*Department of Chemistry, Peking University*

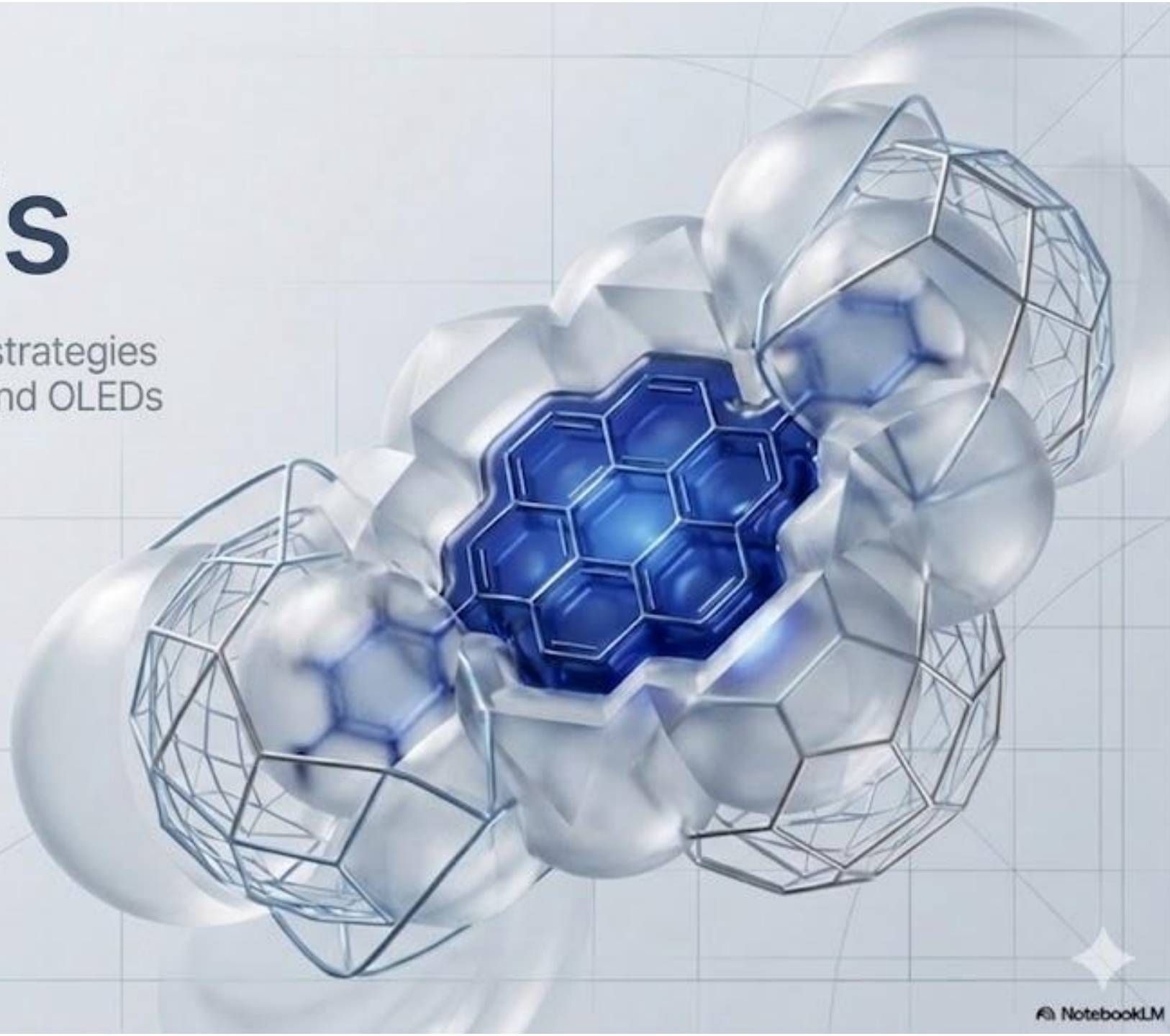
# *Selected Weekly Literature Presentations*

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# MR-TADF<sub>s</sub>

Systematic structural engineering strategies to unlock high-efficiency, narrowband OLEDs at mass-production doping ratios.



Zisheng Xue  
2026.04.11

# ***MR-TADF*s**

## ***Content***

- *TADFs and Essential Theoretical Backgrounds*
- *D-A type TADFs and their limitations*
- *MR-TADFs*
- *Towards Deep RED Emission*
- *Towards Brighter OLEDs: faster RISC*
- *Towards Brighter OLEDs: against ACQs*

# MR-TADFs

**Multiple Resonance**

***Thermally Activated  
Delayed Fluorescence***



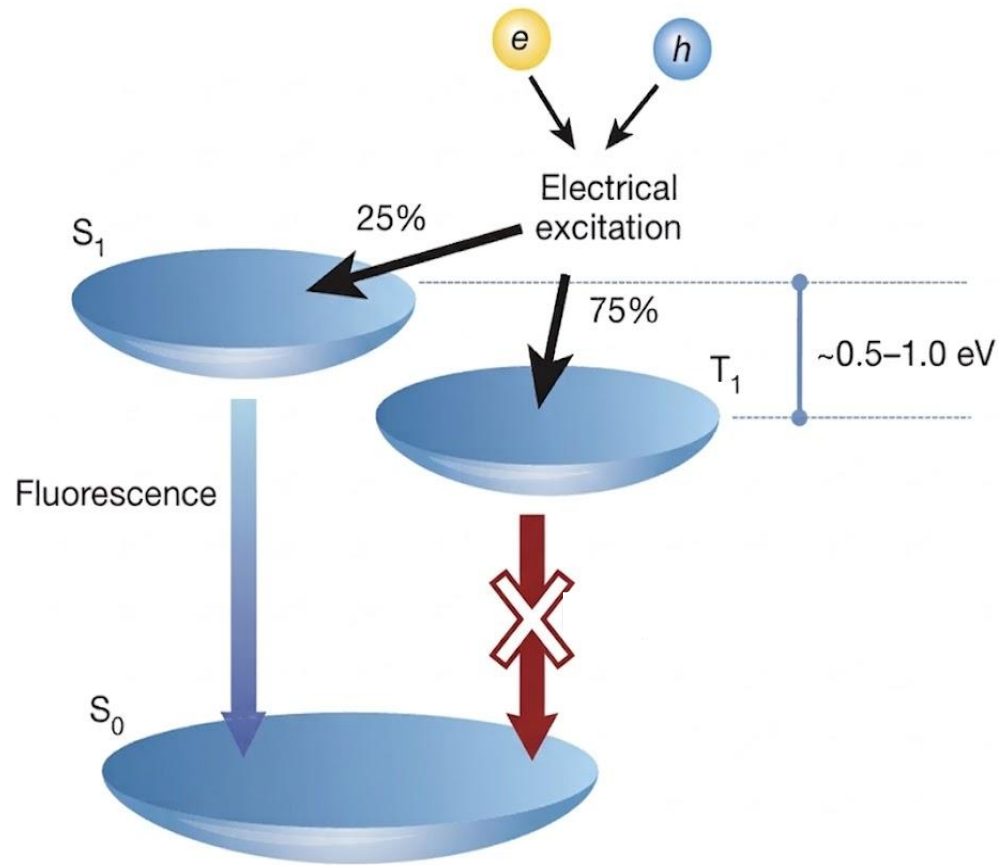
"I recognize the  
words, but **NOT**  
the whole  
sentence

***History and Essential theoretical  
background of TADF***

# What is TADF and Why we need TADF?

When a device is electrically excited: 3:1 ratio of T:S

For Normal fluorescent material **only S state** can be used for **emitting lights**: **Maxium EQE: 25%**

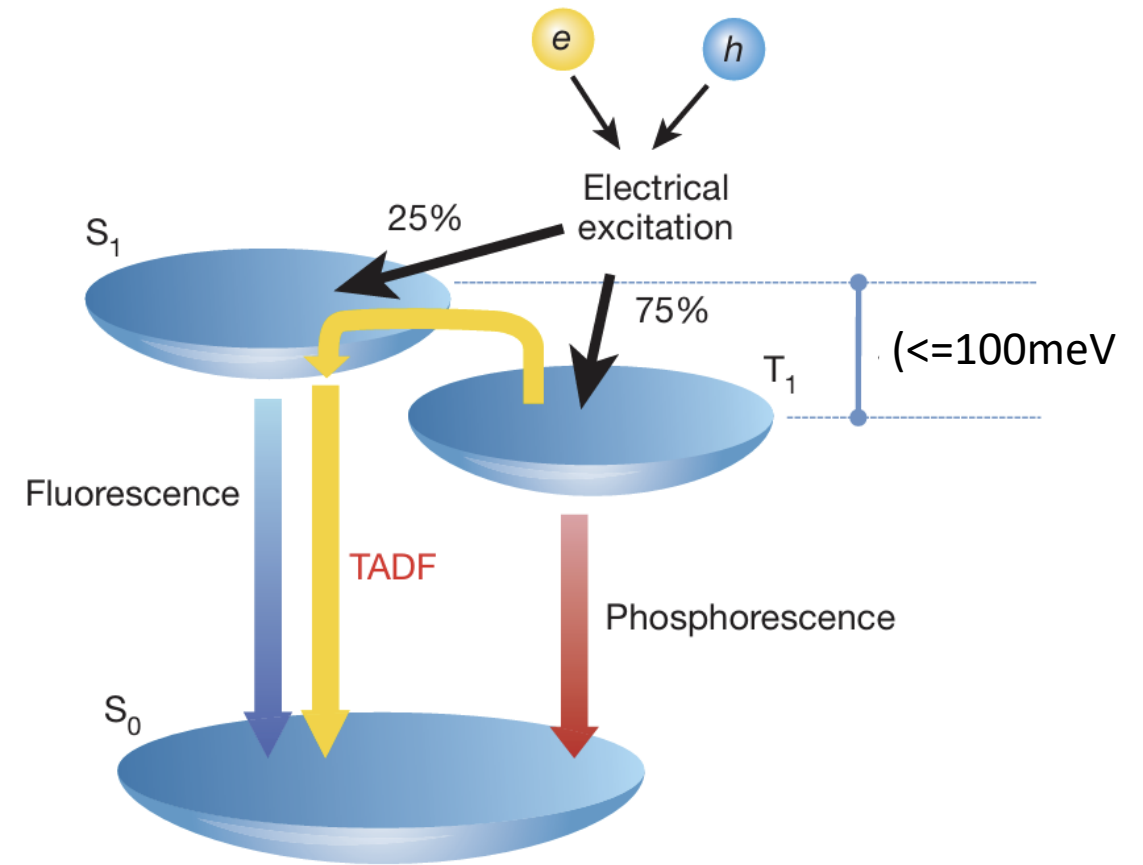


$\Delta E_{ST}$  small enough ( $\leq 100\text{meV}$ )



**Maxium EQE: 100%**

**Fast enough RISC (Reverse ISC, T1→S1)**



# What determines $\Delta E_{ST}$

$$\Delta E_{\{ST\}} = 2J$$

$$J = \iint \phi_H(r_1)\phi_L(r_1) \frac{e^2}{|r_1 - r_2|} \phi_H(r_2)\phi_L(r_2) d\mathbf{r}_1 d\mathbf{r}_2$$

Exchange Integral  
(交换积分)

Physical Meaning:

泡利不相容原理，三线态的两个电子无法在空间中靠近。减少了电子斥力

$$\Delta E_{ST} \propto K \propto \langle \phi_H | \phi_L \rangle$$

HOMO-LUMO overlap

Molecular design?



Small HOMO-LUMO  
overlap



Small  $\Delta E_{ST}$



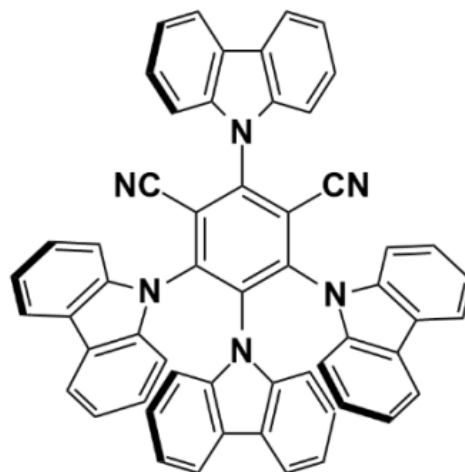
Faster RISC; better EQE

# Gen1 TADF molecular Donor- Acceptor type

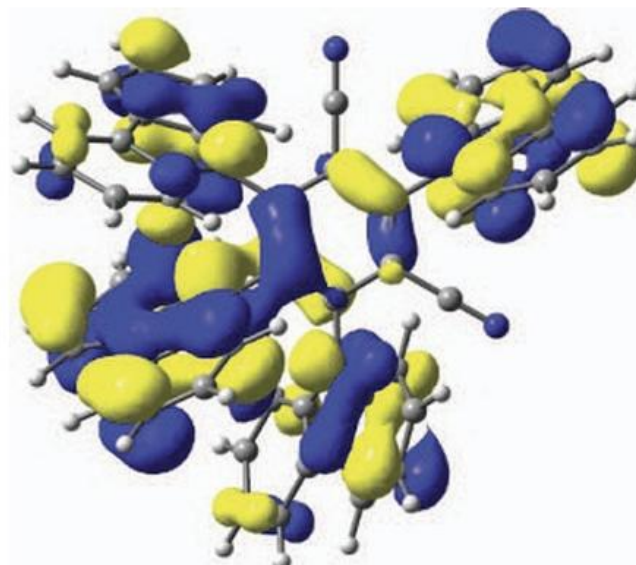


Chihaya Adachi (安達千波矢)

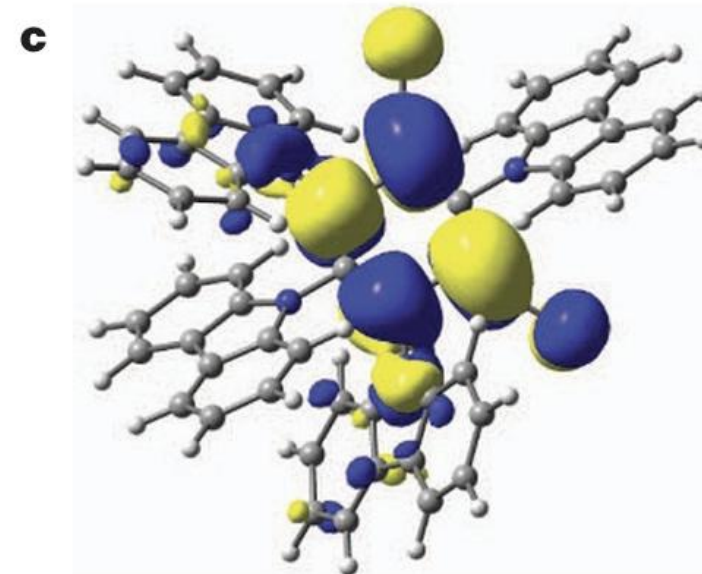
Kyushu University



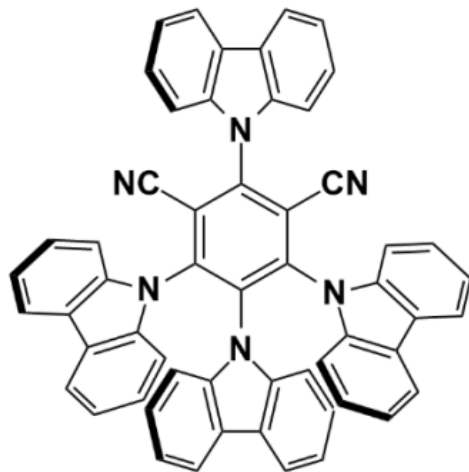
4CzIPN



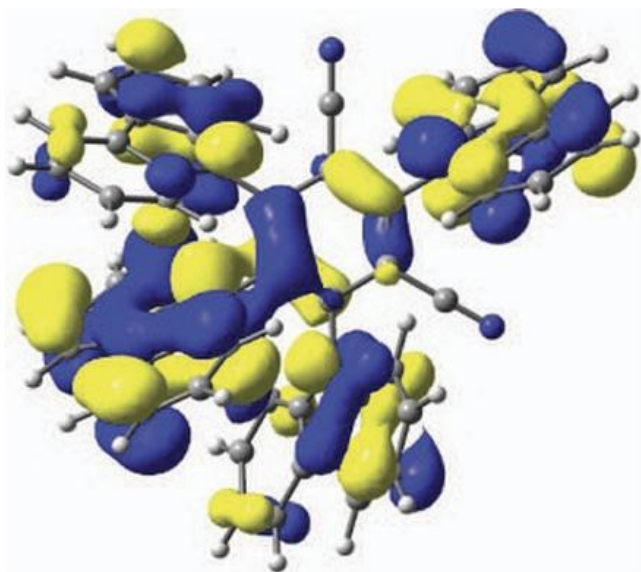
Highly distorted from the dicyanobenzene plane by steric hindrance  
HOMO/LUMO locates on Donor/Acceptor  
S1 usually represents a Charge transfer state  
(donor to acceptor)



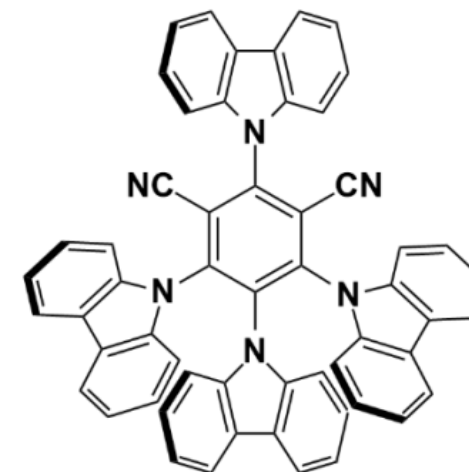
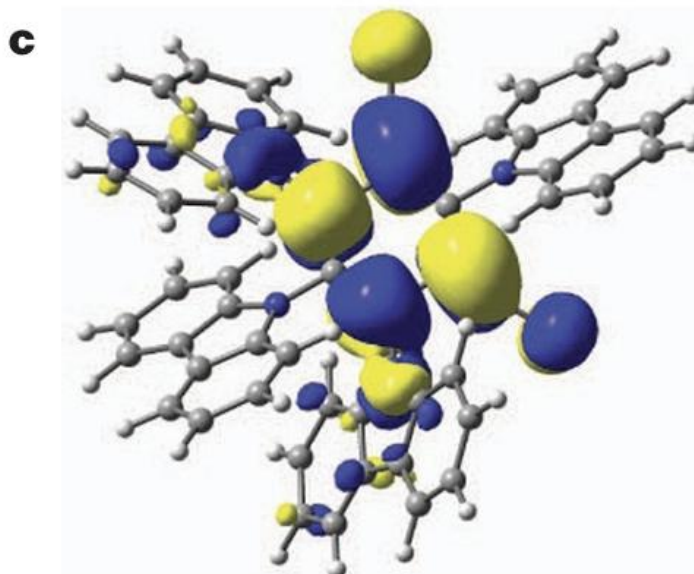
# Gen1 TADF molecular Donor- Acceptor type



4CzIPN



Highly distorted from the dicyanobenzene plane by steric hindrance  
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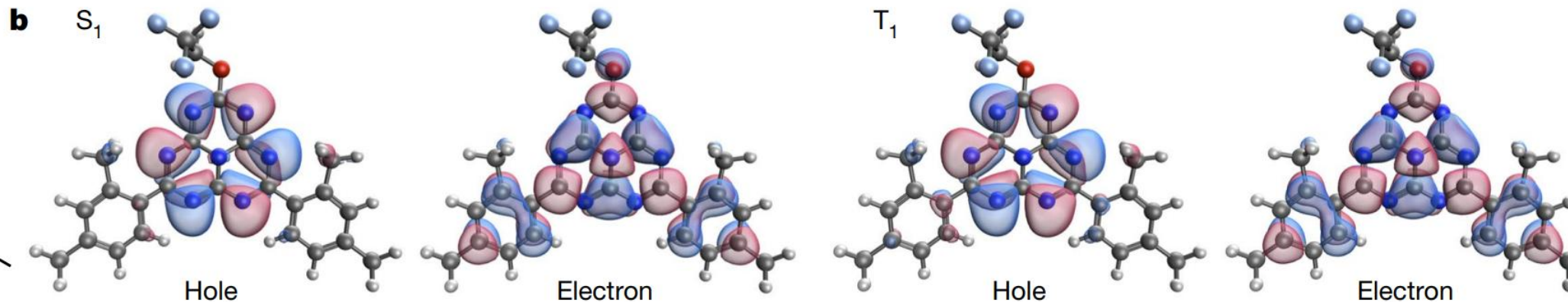
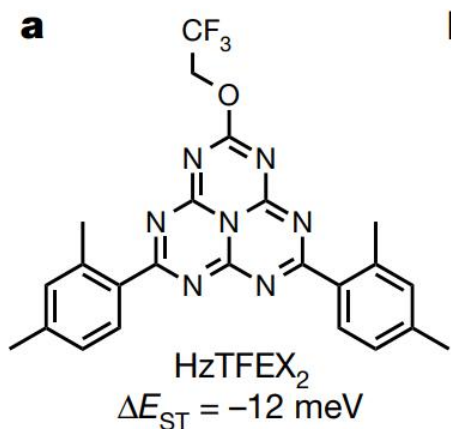


4CzIPN

$\Delta E_{ST} = 83\text{meV}$   
EQE=11.2%  
Much higher than 1gen OLED  
(apox. 5%)

# How small $\Delta E_{ST}$ could be?

$\Delta E_{ST}$  could be  $<0$  with proper molecular design



But, At what cost?

$$f \propto \left| \int \phi_H(r) r \phi_L(r) dr \right|^2$$

振子强度:  
描述发光  
“速率”

For HzTFEX<sub>2</sub> only very low emission

# What determines $k_{RISC}$

$\Delta E_{ST}$  is NOT the only factor matters in RISC process

Spin-Orbit Coupling  $\searrow$

$$k_{RISC} \propto |H_{SOC}|^2 \cdot \exp\left(-\frac{\Delta E_{ST}}{k_B T}\right)$$

$$\widehat{H}_{SOC} = \sum_i \xi(r_i) \widehat{L}_i \cdot \widehat{S}_i$$

**Both  $S_1$  and  $T_1$  are Charge transfer state (usually  $\pi-\pi$ )** “Heavy atom effect “

**Other state need to be coupled**

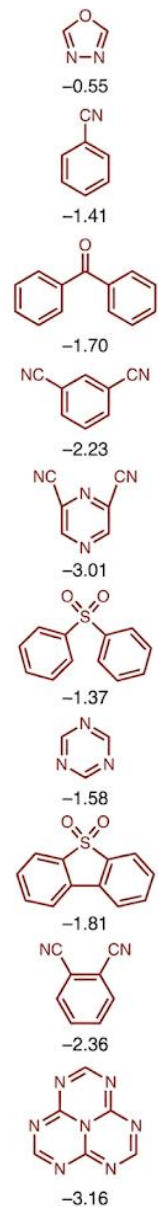
**Normally it should be  ${}^3LE$**

$$\langle S_1 | \widehat{H}_{SO} | T_1 \rangle_{eff} = \frac{\langle {}^1CT | \widehat{H}_{SO} | {}^3LE \rangle \langle {}^3LE | \widehat{H}_{vib} | {}^3CT \rangle}{\Delta E(CT - LE)}$$

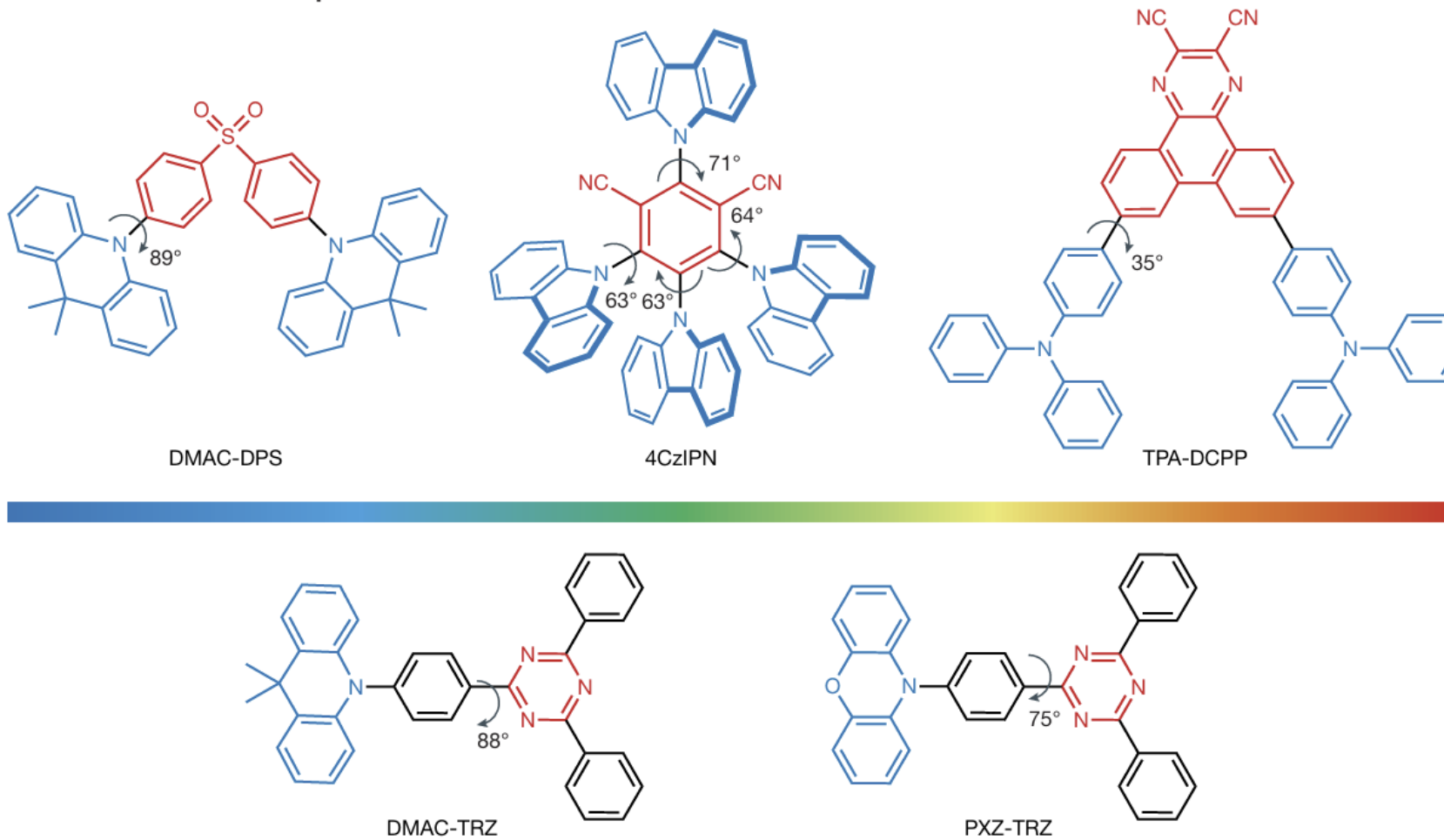
**For Acceptor/Donor Design an Energy Accessible LE State Should be Find**

# Donor- Acceptor type TADF materials

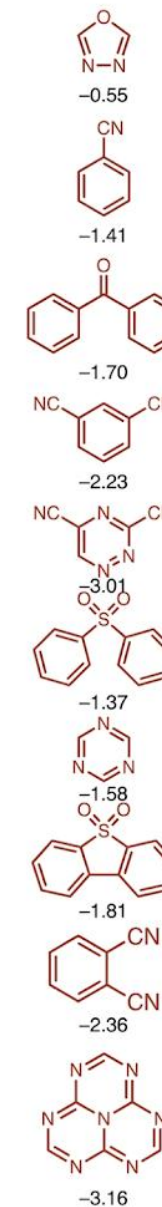
Acceptor (LUMO, eV)



Chemical structures of representative TADF molecules



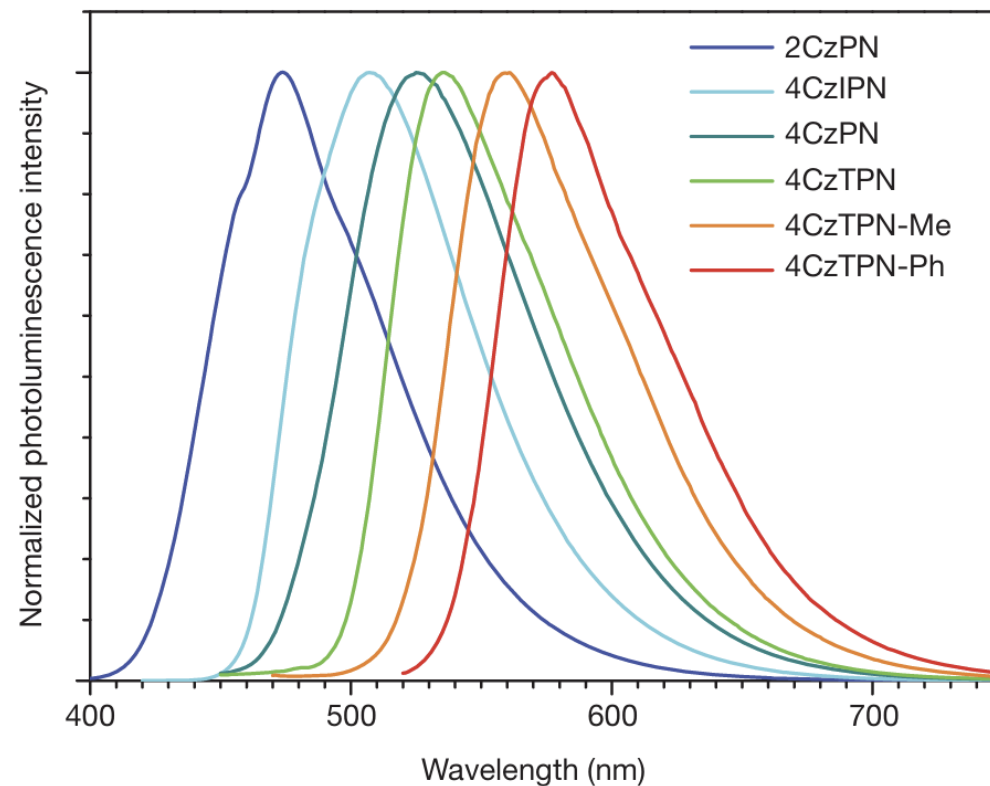
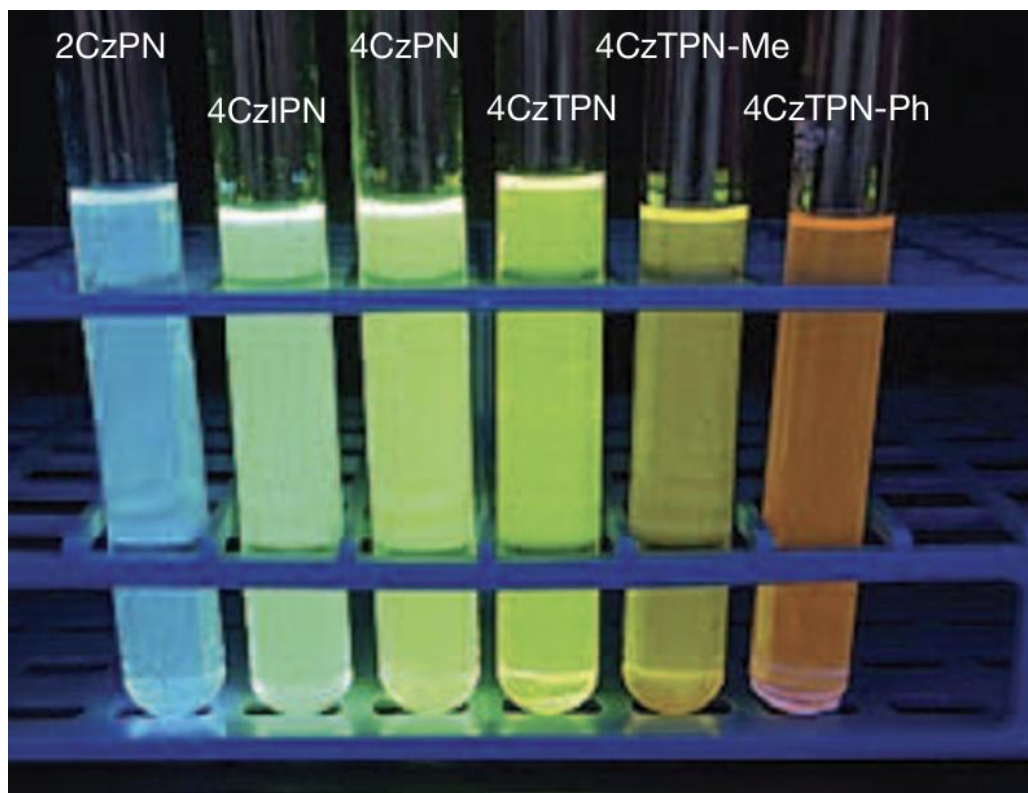
Acceptor (LUMO, eV)



# Major Drawbacks in Gen1 TADF

Several drawbacks (low stability, roll-off, etc.) origins from the D-A molecular

But there's one that is Intolerable.



No way this is a blue emitter.

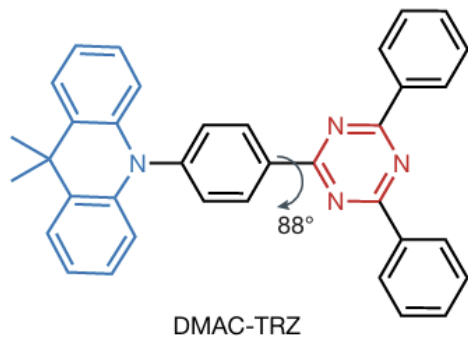
# What determines FWHM?

$$FWHM = 2\sqrt{2 \ln 2}$$

$$\sigma^2 = \sum_{\alpha} S_{\alpha} (\hbar\omega_{\alpha})^2 (2n_{\alpha} + 1)$$

$$S_{\alpha} = \frac{M_{\alpha} \omega_{\alpha} \Delta Q_{\alpha}^2}{2\hbar}$$

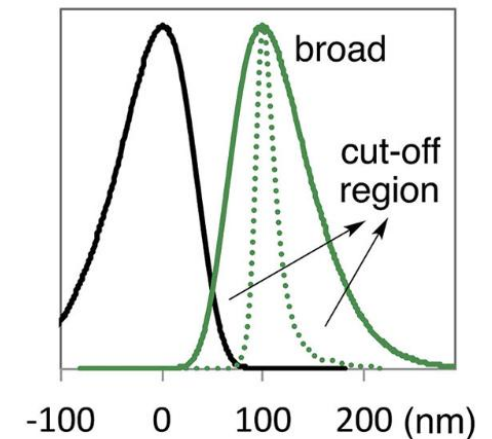
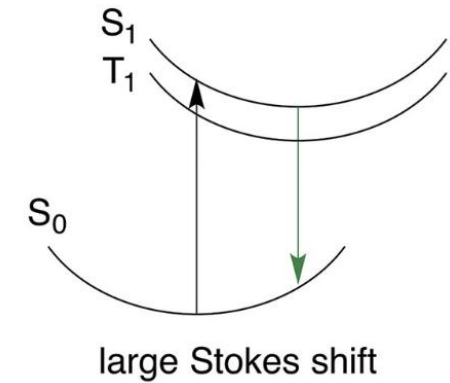
$\Delta Q_{\alpha}$ : measurement of structure difference between Excite state and ground state.



S1 of D-A type TADF: CT state huge difference

How to achieve small structure difference ?

Simplified graphical explanation



# ***MR-TADF*s**

## ***Content***

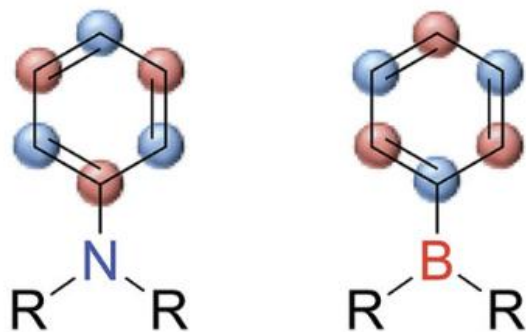
- *TADF*s and Essential Theoretical Backgrounds
- *D-A* type *TADF*s and their limitations
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# MR-TADF

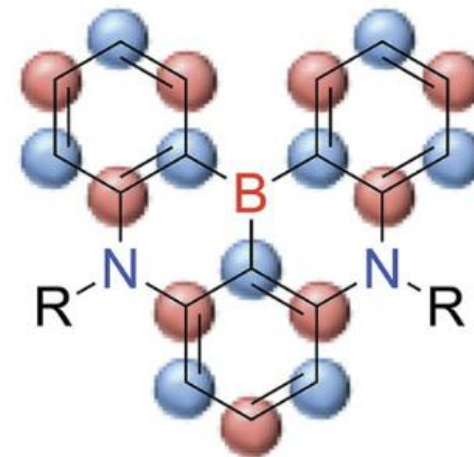


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Takuji Hatakeyama

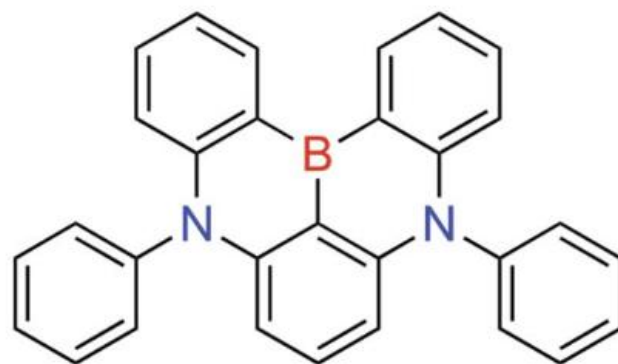
new TADF molecule



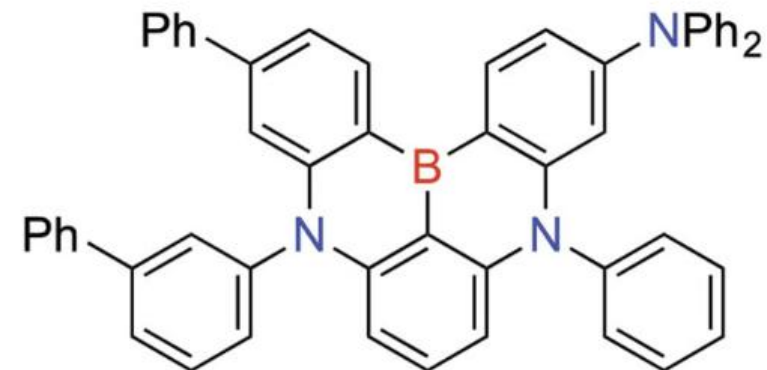
opposite resonance effect



HOMO-LUMO separation  
by multiple resonance effect



**DABNA-1**

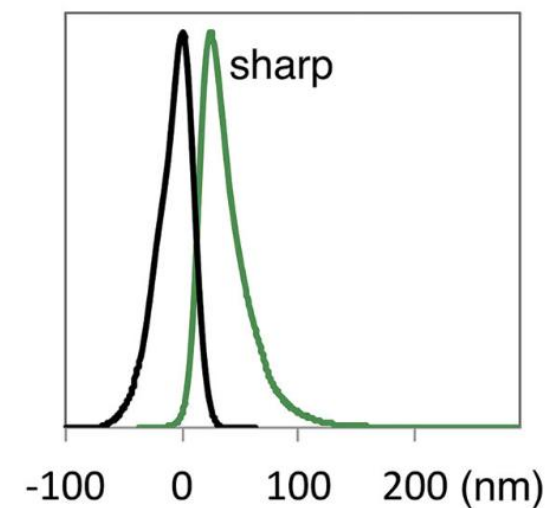
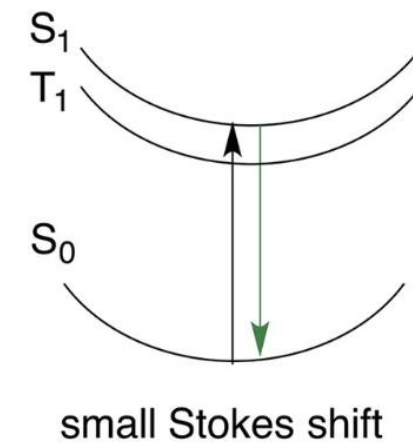
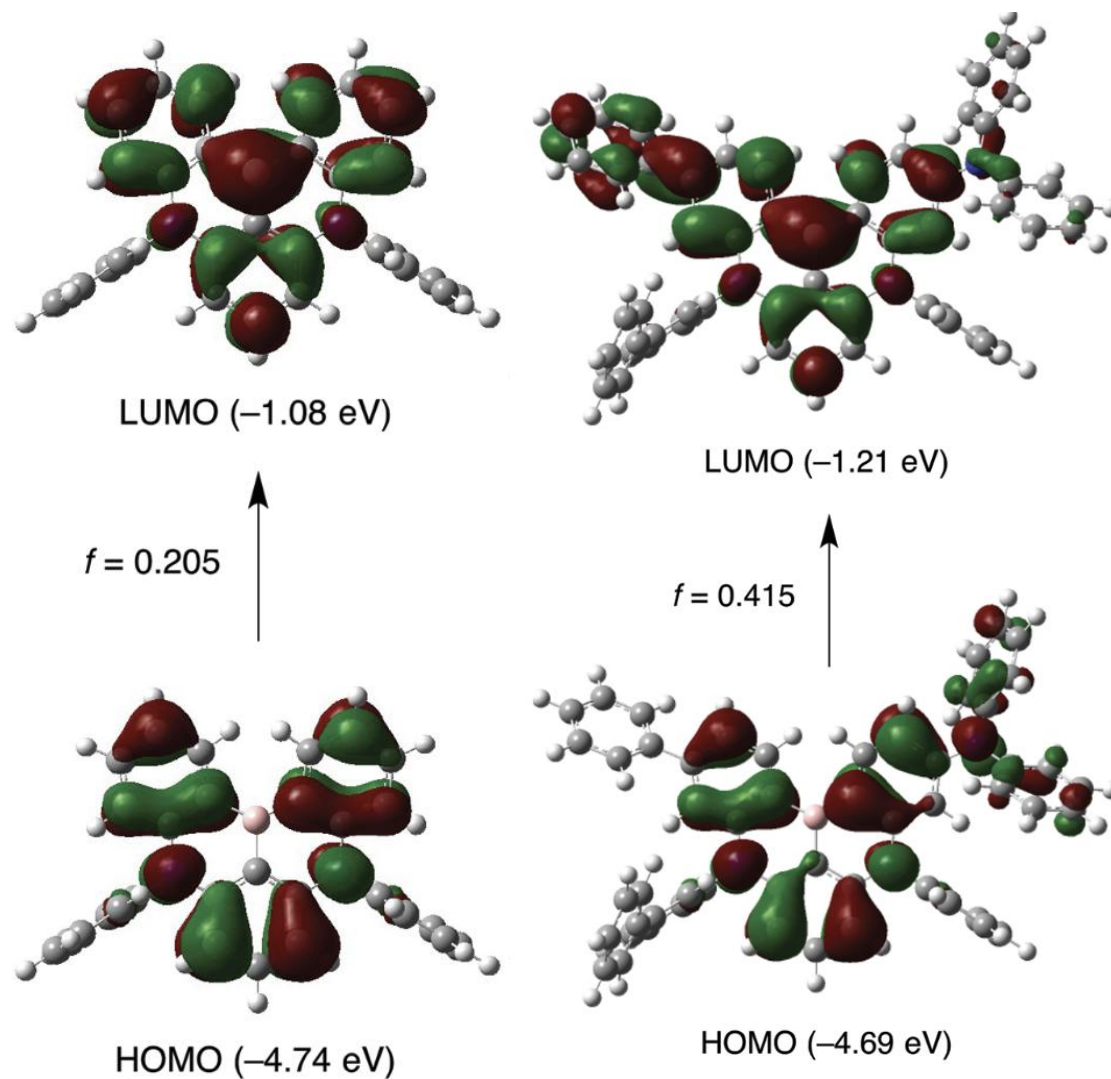


**DABNA-2**

# MR-TADF

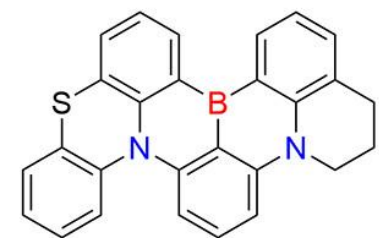


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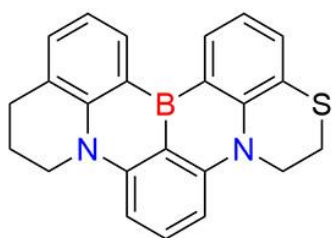


# Three major family of MR-TADF Molecular

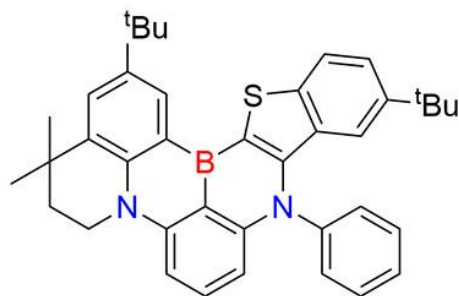
## B-N type molecular



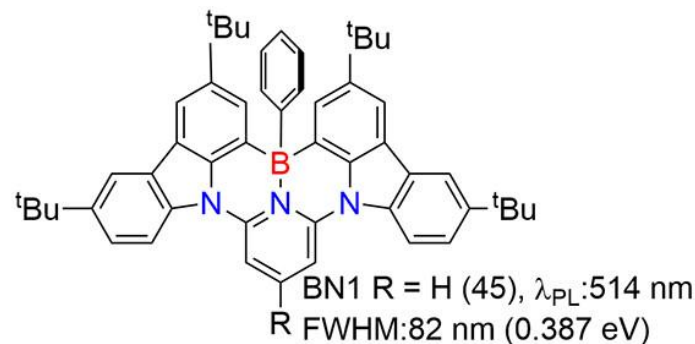
B-N- S-1 (42),  $\lambda_{\text{PL}}$ :483 nm  
FWHM:41 nm (0.218 eV)



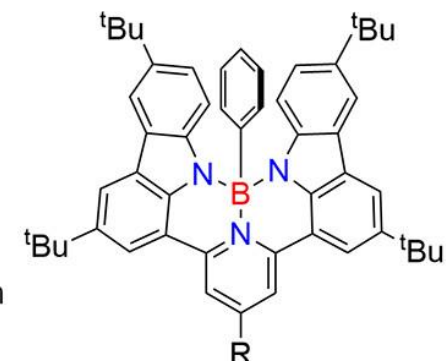
B-N- S-2 (43),  $\lambda_{\text{PL}}$ :458 nm  
FWHM:33 nm (0.195 eV)



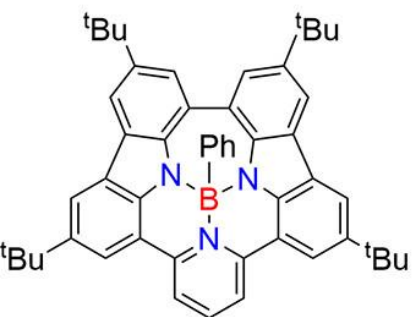
B-N- S-3 (44),  $\lambda_{\text{PL}}$ :457 nm  
FWHM:25 nm (0.149 eV)



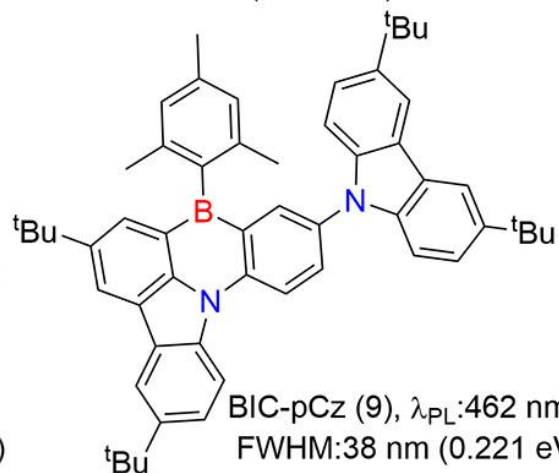
TCz-BN1 R = tCz (46),  $\lambda_{\text{PL}}$ :517 nm  
FWHM:89 nm (0.416 eV)



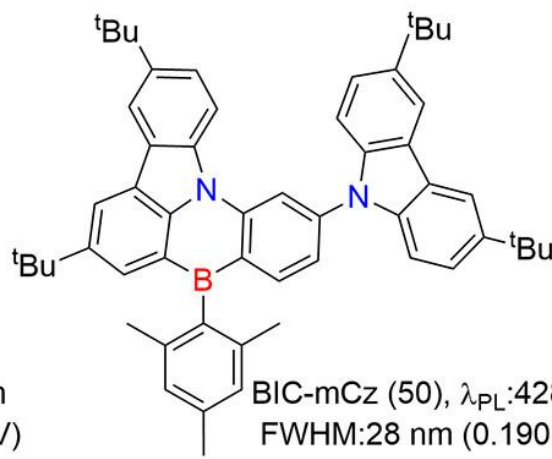
BN2 R = H (47),  $\lambda_{\text{PL}}$ :567 nm  
FWHM:97 nm (0.377 eV)  
TCz-BN2, R = tCz (48),  $\lambda_{\text{PL}}$ :584 nm  
FWHM:108 nm (0.396 eV)



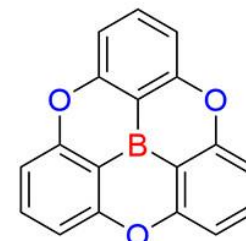
BN3 (49),  $\lambda_{\text{PL}}$ :694 nm  
FWHM:151 nm (0.393 eV)



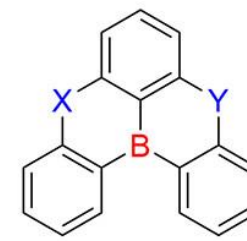
BIC-pCz (9),  $\lambda_{\text{PL}}$ :462 nm  
FWHM:38 nm (0.221 eV)



BIC-mCz (50),  $\lambda_{\text{PL}}$ :428 nm  
FWHM:28 nm (0.190 eV)



BOOO (51)  
 $\lambda_{\text{PL}}$ :386 nm



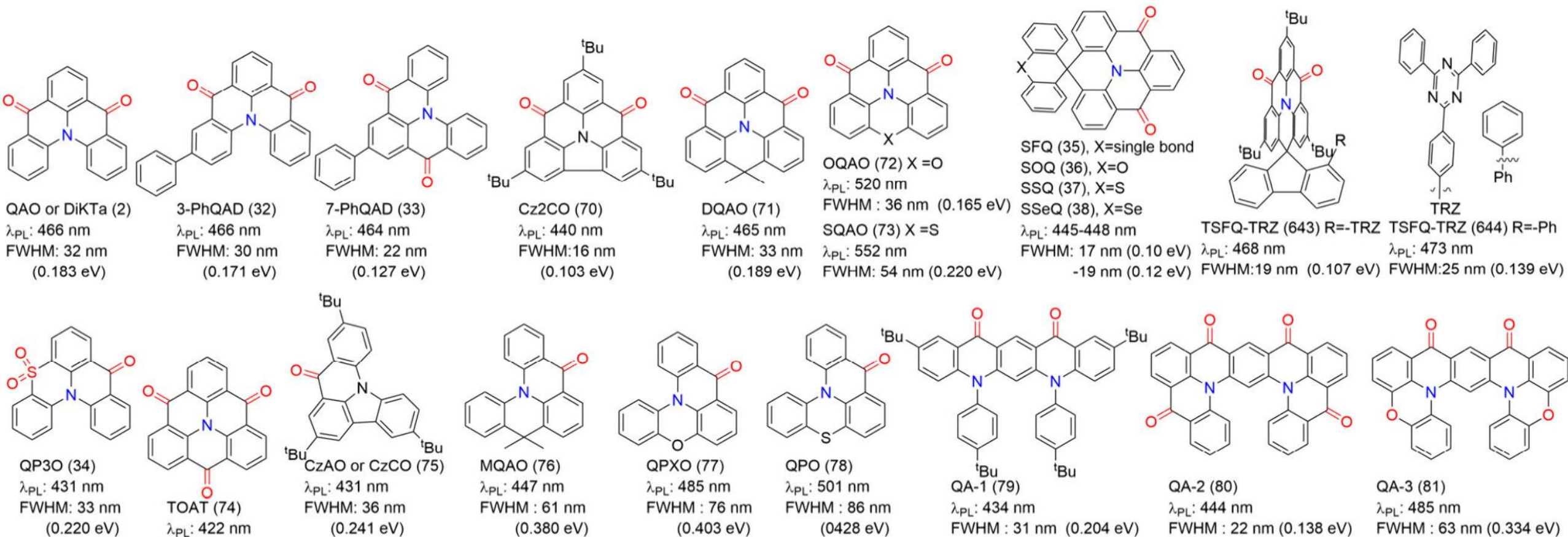
BOO X = Y = O or DOBNA (52)  
 $\lambda_{\text{PL}}$ :396 nm, FWHM:30 nm (0.238 eV)

BOS X=O Y= S (53),  $\lambda_{\text{PL}}$ :434 nm  
FWHM:29 nm (0.191 eV)

BSS X=S Y= S (6),  $\lambda_{\text{PL}}$ :457 nm  
FWHM:27 nm (0.160 eV)

# Three major family of MR-TADF Molecular

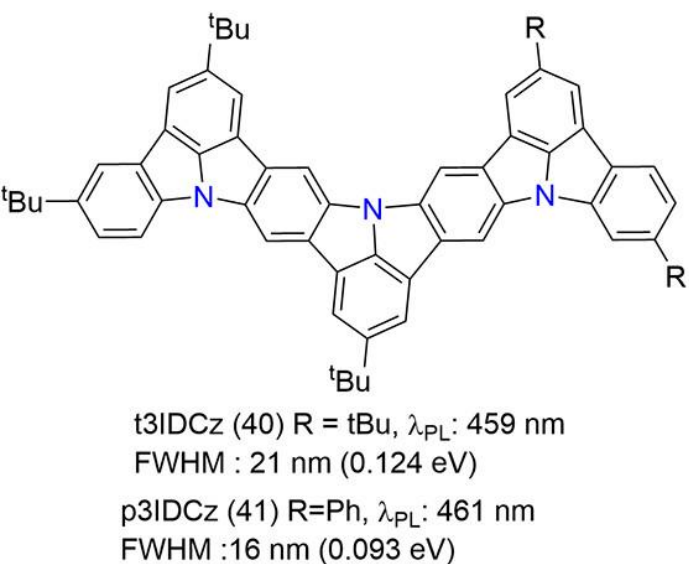
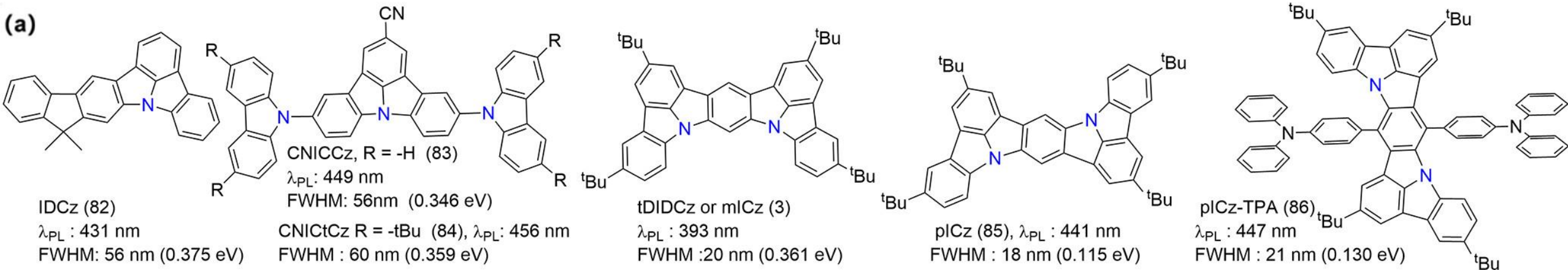
## N-CO type molecular



MR properties relays on CO's pair\*

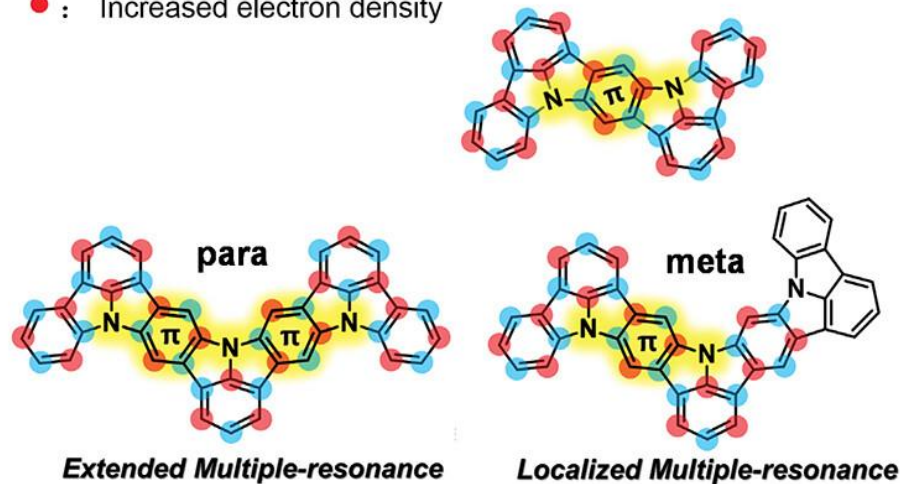
# Three major family of MR-TADF Molecular

## N-PAHs type molecular



### (b) N- $\pi$ -N Extension

- : Decreased electron density
- : Increased electron density



MR properties relays on PAH's  $\pi$ ai\*

# ***MR-TADF*s**

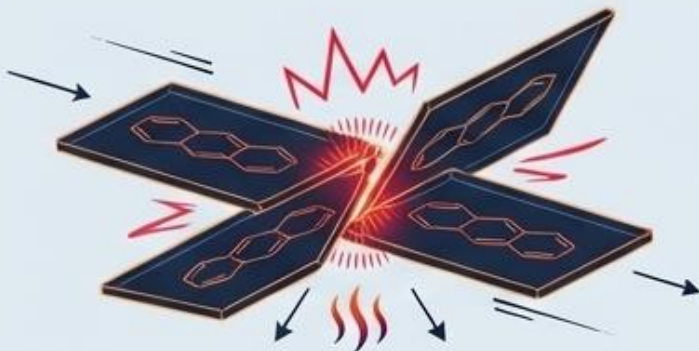
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- *TADFs and Essential Theoretical Backgrounds*
- *D-A type TADFs and their limitations*
- *MR-TADFs*
- *Towards Deep RED Emission*
- *Towards Brighter OLEDs: faster RISC*
- *Towards Brighter OLEDs: against ACQs*

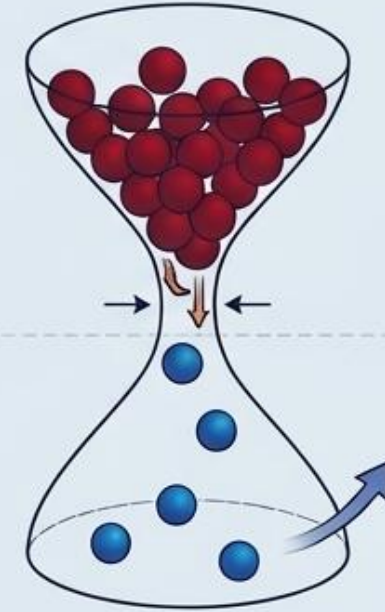
# Three Key Challenges within MR-TADFS



**Full-Color, Narrowband,  
High-Efficiency  
Electroluminescence**



**Avoiding ACQ  
In solid phase: breaking the  
Aggregation through molecular  
design**



**Avoiding roll-offs and TTAs:  
Towards faster  $K_{IST}$**

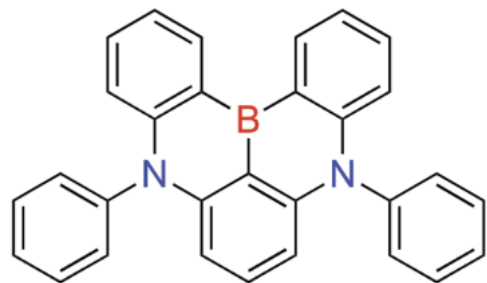
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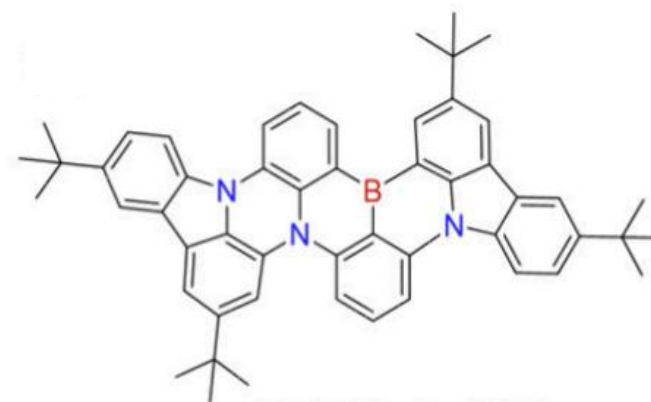
# Towards Deep RED Emission: The issue

Challenge: Adding Donor/Acceptor on the compound  
Will cause decrease property in MR-TADF



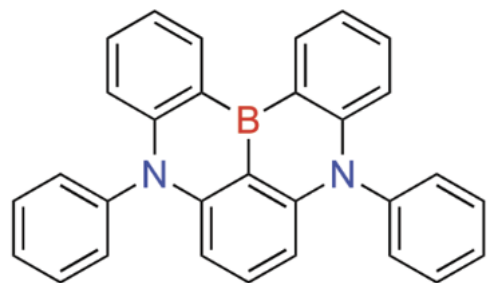
**DABNA-1**

$\lambda_{\text{PL}}$ : 437 nm  
FWHM: 33 nm



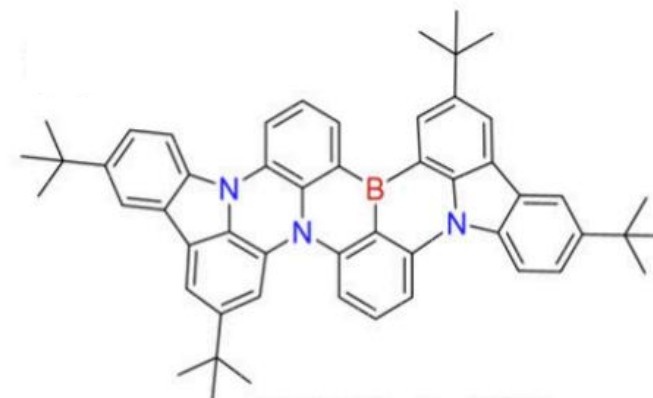
Small step in wavelength  
Huge step in wavewidth

$\lambda_{\text{PL}}$ : 564 nm  
FWHM: 60 nm



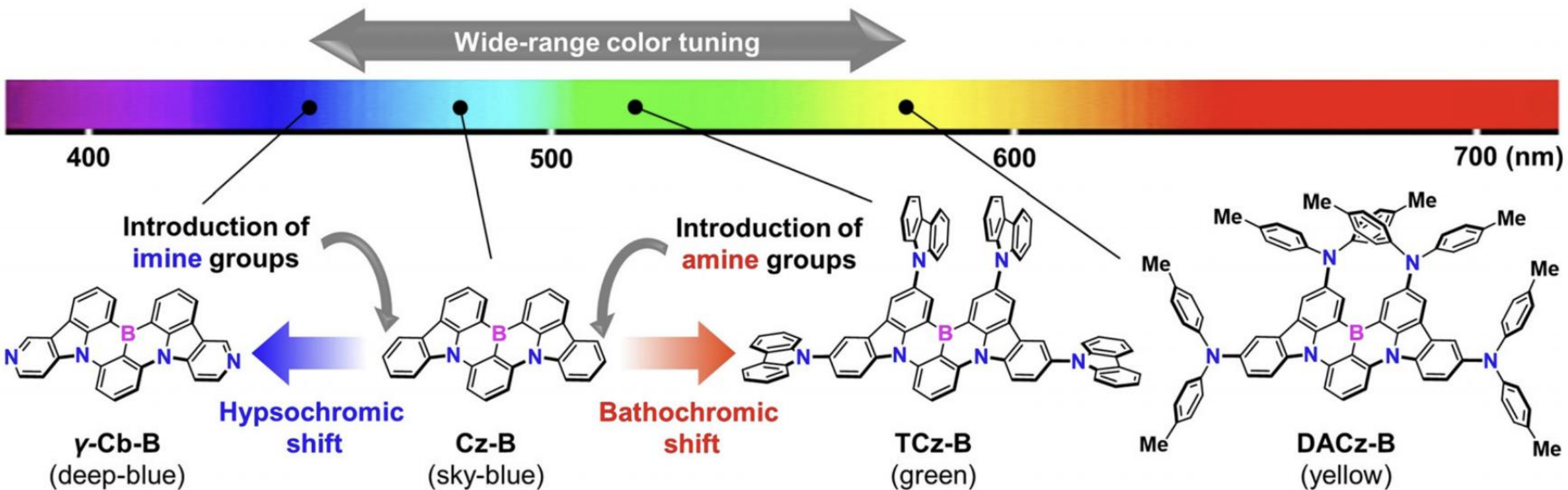
**DABNA-1**

Short-Term Charge  
Transfer Dominates (SRCT)

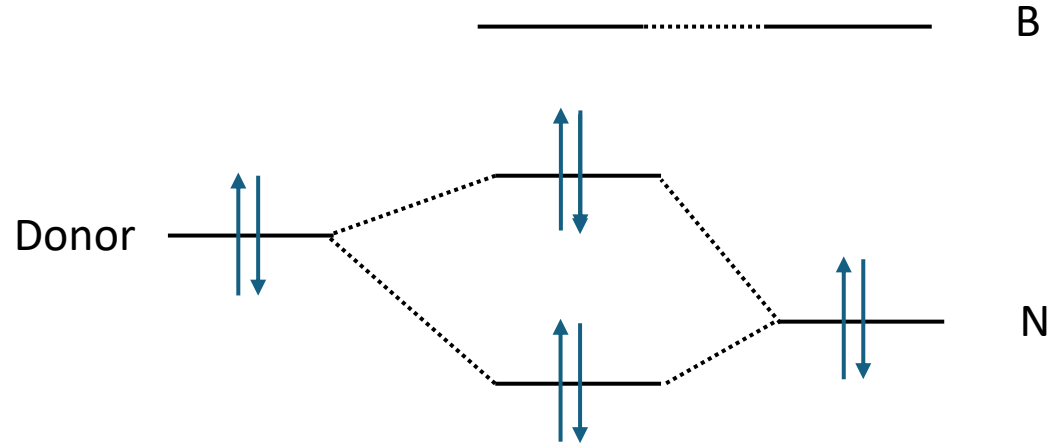


More **LRCT** combines  
More properties like D-A type TADFs

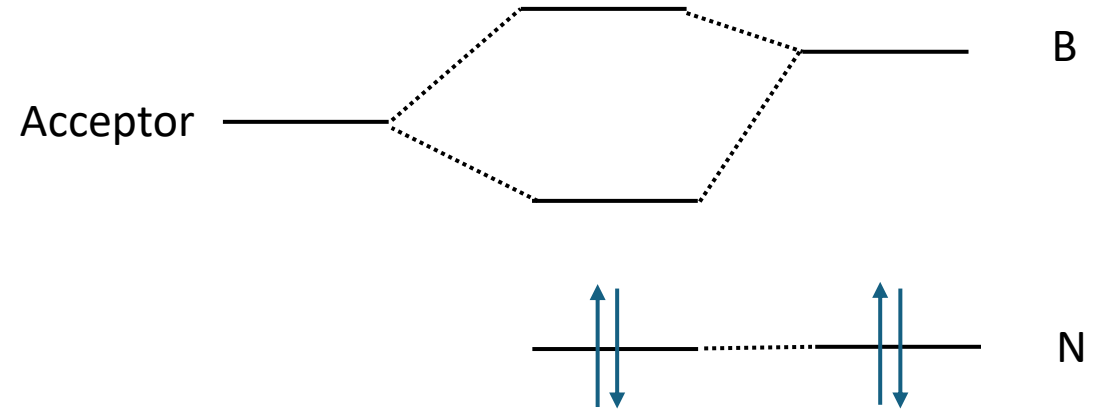
# Solution 1: Finding the balance



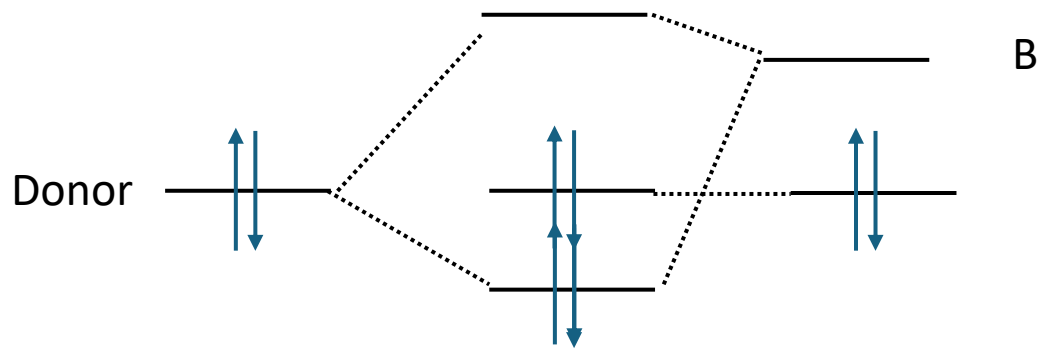
# A-para-B/ D-para-N



D-para-N: Red shift



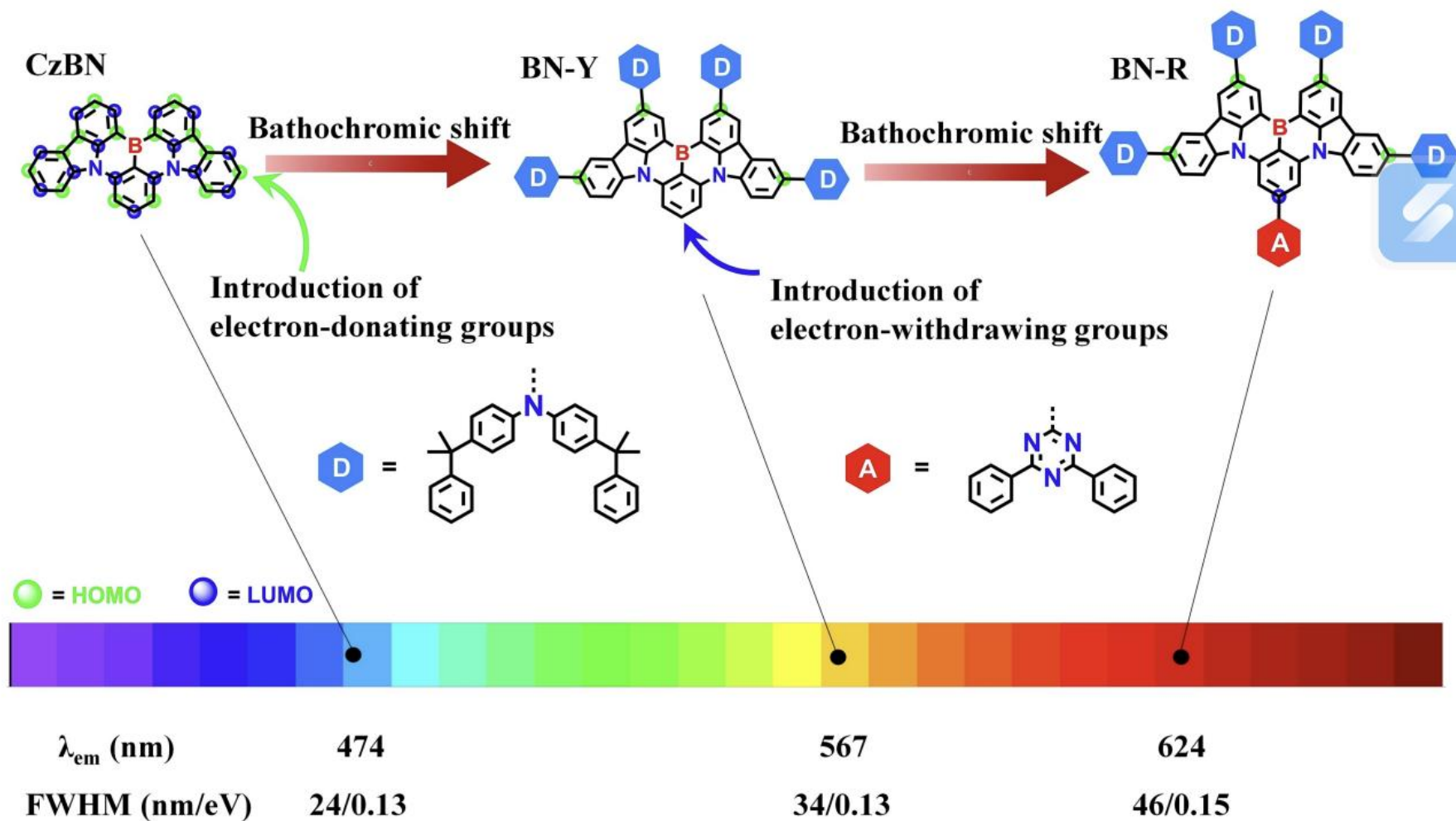
A-para-B: Red shift



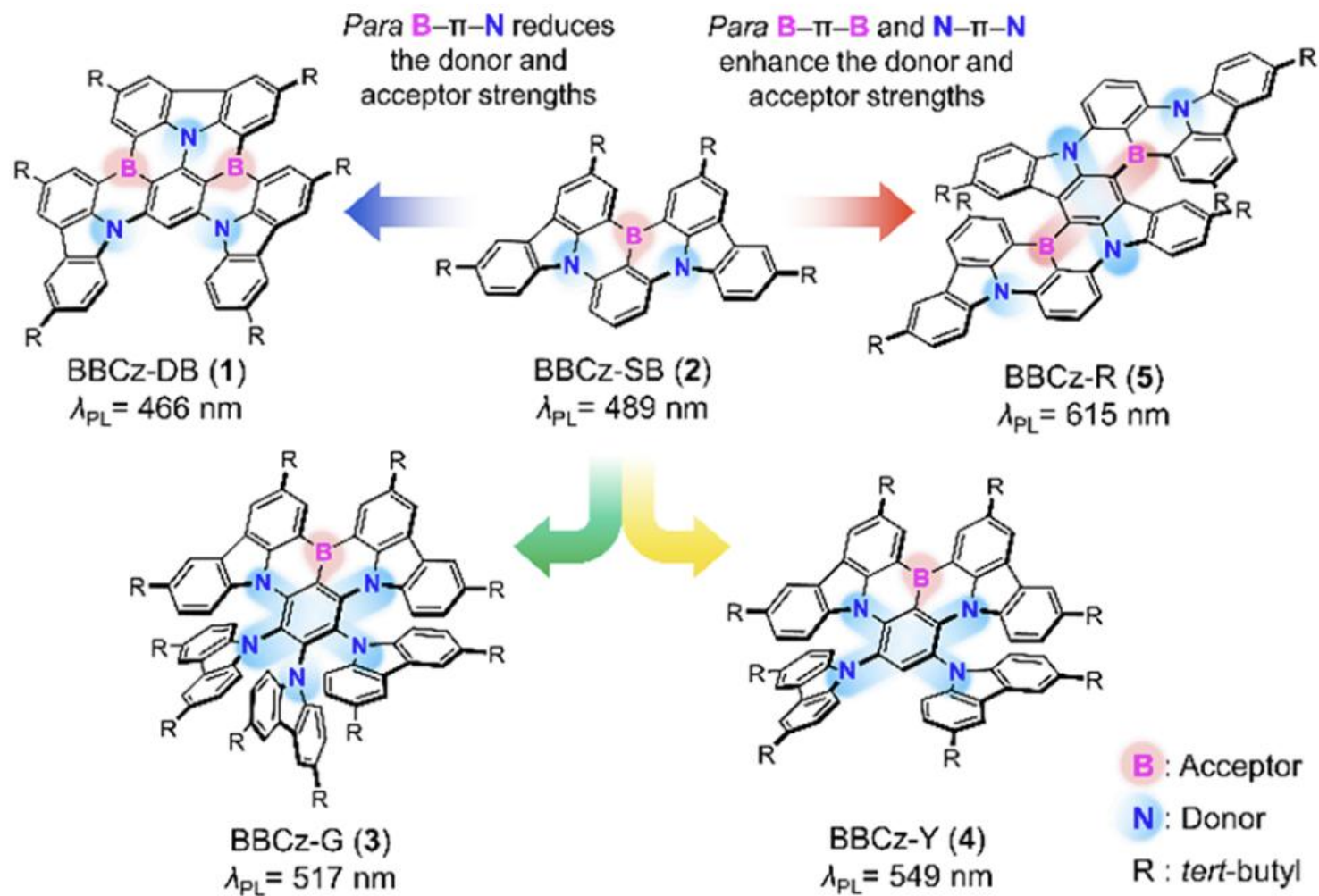
D-para-B: Blue Shift

*And vice versa*

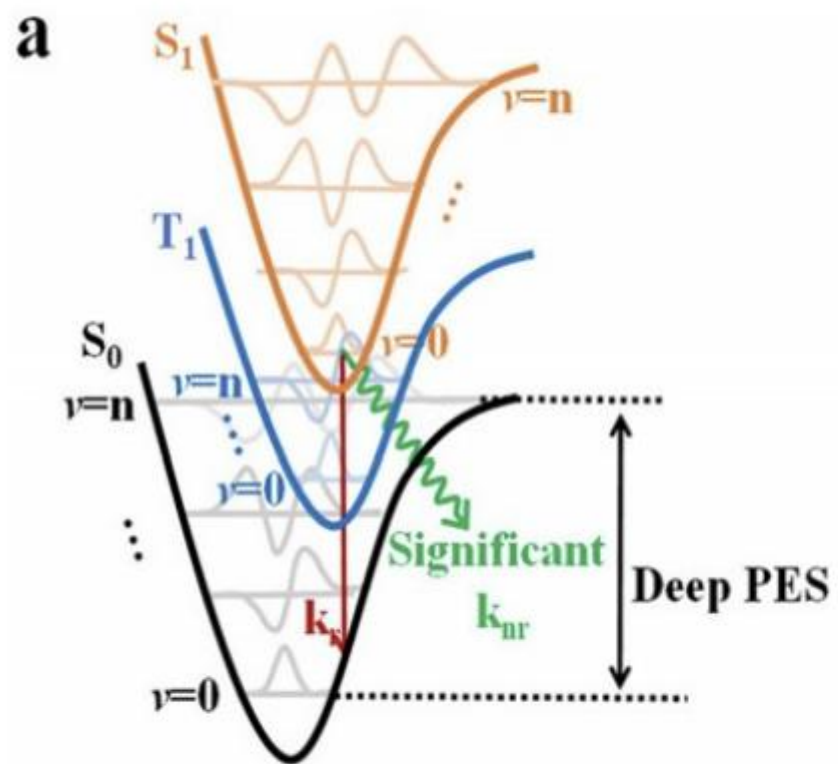
# Solution 1: Finding the balance



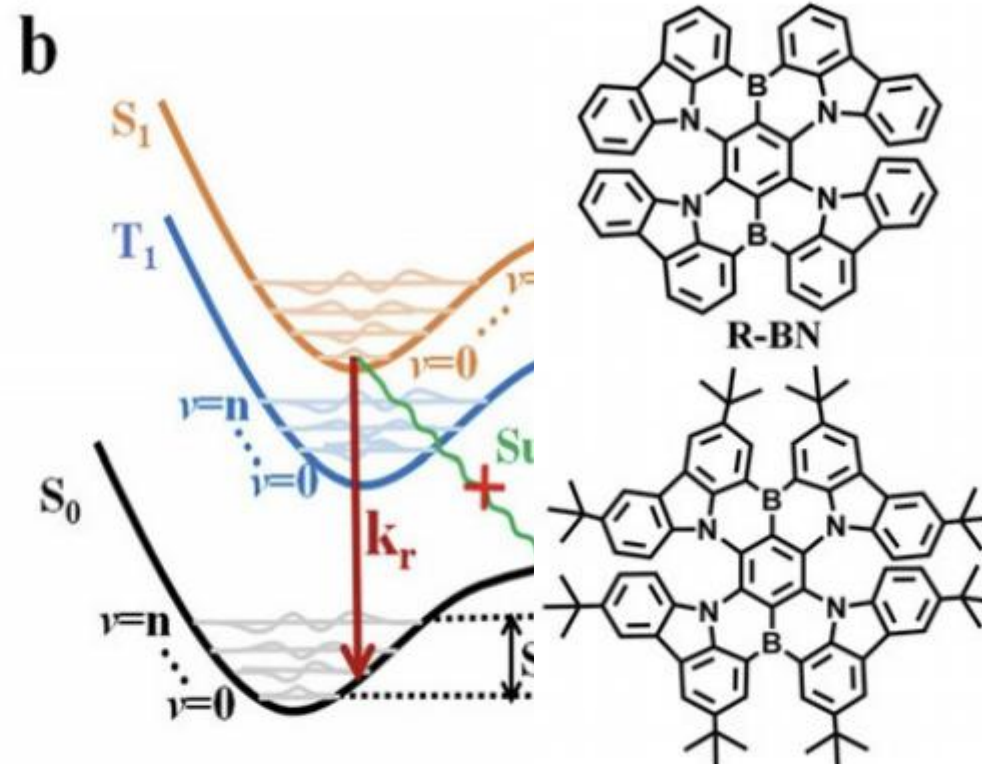
# Solution 2: Enhancing SRCT/LRCT



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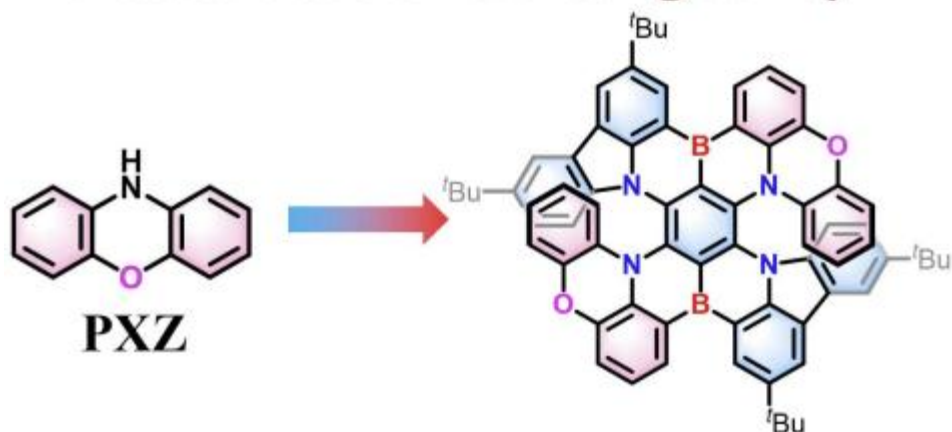
Strong  $S_1$ - $S_0$  vibrational coupling



Weak  $S_1$ - $S_0$  vibrational coupling

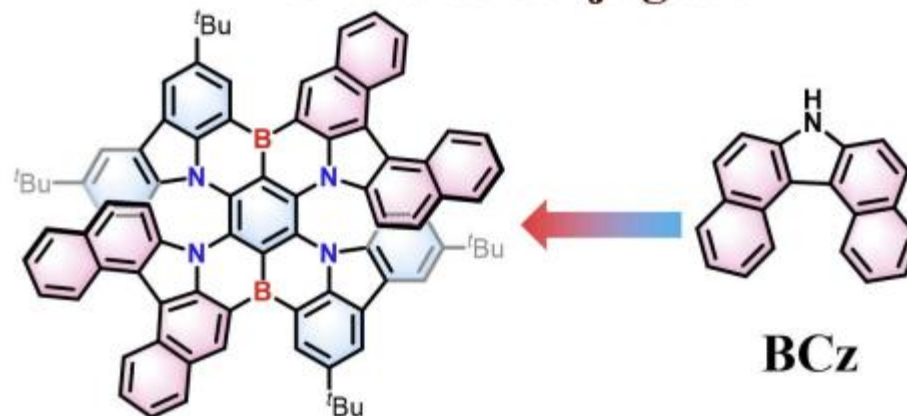
# Solution 2: Enhancing SRCT/LRCT

Increased electron-donating ability



$\lambda_{\text{PL}} = 697 \text{ nm}$   
FWHM = 55 nm (0.14 eV)

Extended  $\pi$ -conjugation



$\lambda_{\text{PL}} = 718 \text{ nm}$   
FWHM = 52 nm (0.12 eV)

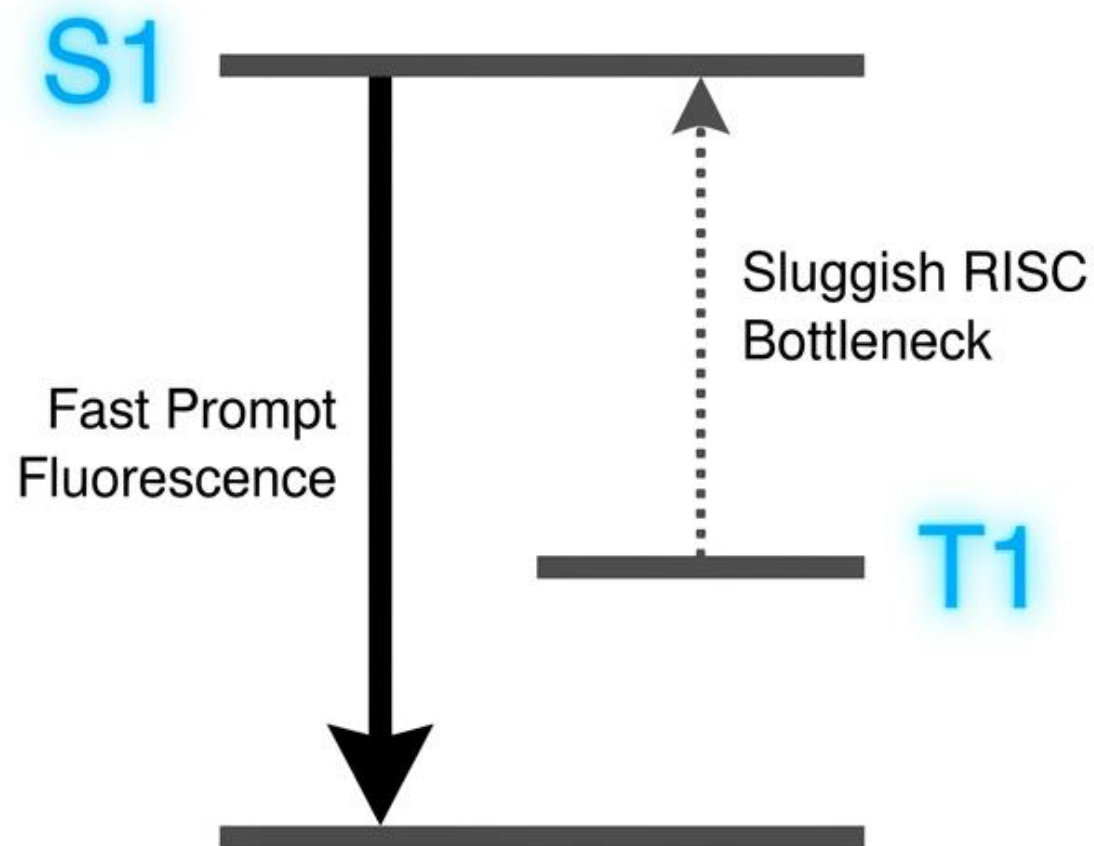
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- *Towards Brighter OLEDs: against ACQs*
- *Beyond MR-TADFs: state-of-the-art*

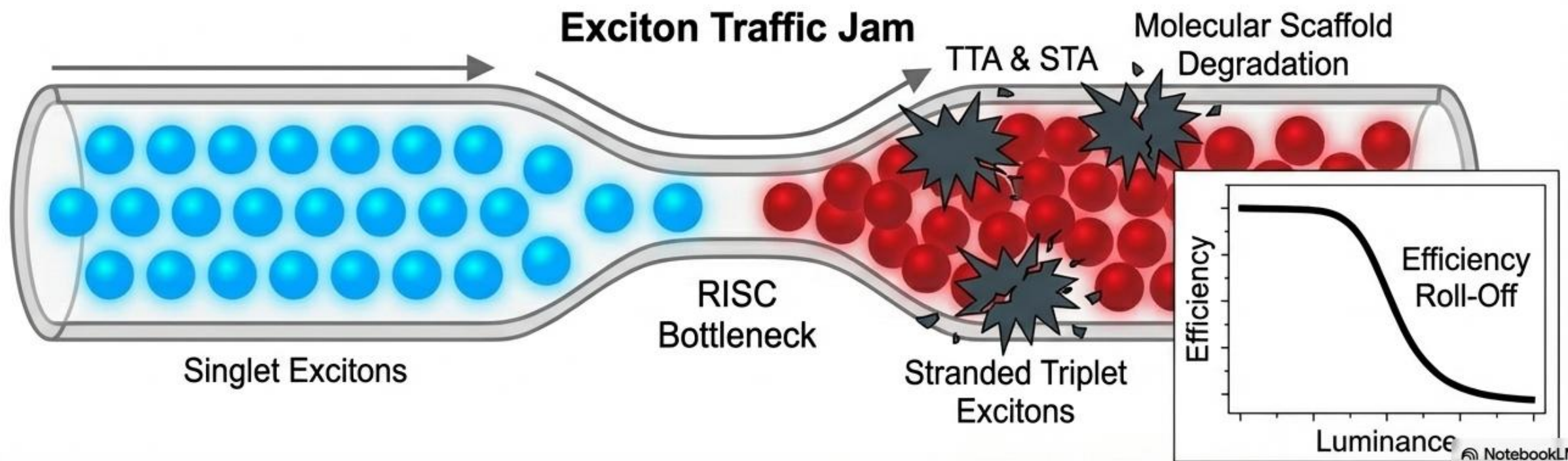
# The Kinetic Bottleneck: Sluggish Reverse Intersystem Crossing

- MR-TADF materials exhibit inherently weak Spin-Orbit Coupling (SOC) between singlet and triplet states.
- This weak SOC results in a severely restricted Reverse Intersystem Crossing rate.



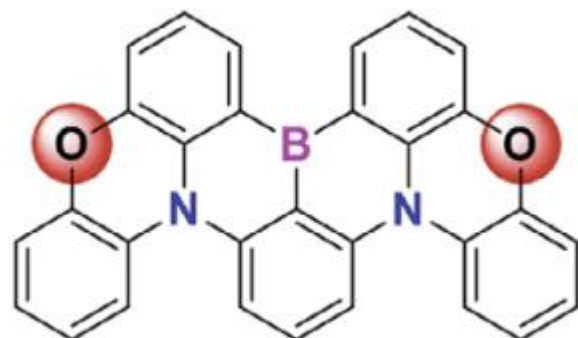
# Consequence: Severe Efficiency Roll-Off and Exciton Annihilation

- At high excitation densities, slow spin-flipping causes massive triplet retention.
- Stranded triplets trigger destructive Triplet-Triplet Annihilation (TTA) and Singlet-Triplet Annihilation (STA).
- Result: Irreversible exciton loss and rapid device aging.

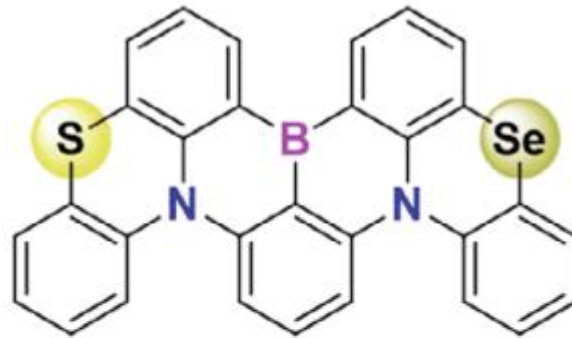


# Solution1: Heavy Atom Effect

Where should we put the heavy atom?



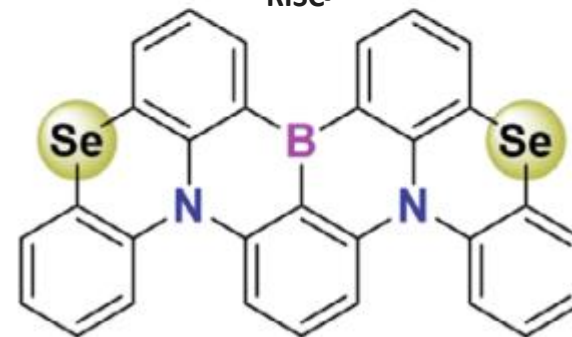
2PXZBN  $k_{\text{RISC}}/10^4=4.3$



BNSSe  $k_{\text{RISC}}/10^4=600$

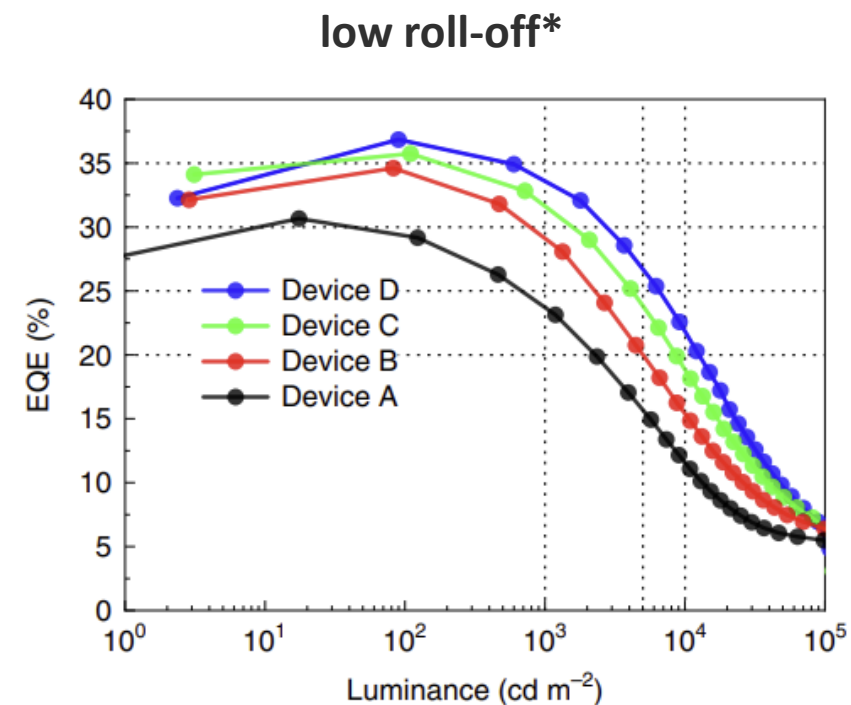


BNSSe  
 $k_{\text{RISC}}/10^4=190$



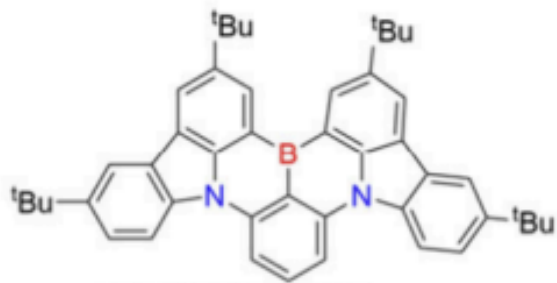
BNSeSe  
 $k_{\text{RISC}}/10^4=2000$

40.5% EQE:  
Super  
dominating

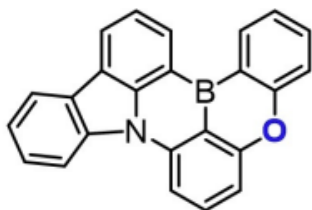


# Solution1: Heavy Atom Effect

Where should we put the heavy atom?

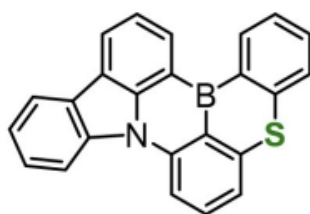


FWHM: 24 nm



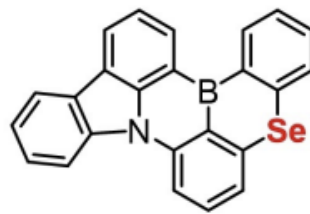
CzBO

FWHM: 26 nm  
 $K_{\text{RISC}}/10^4 = 0.9$



CzBS

FWHM: 28 nm  
 $K_{\text{RISC}}/10^4 = 22$

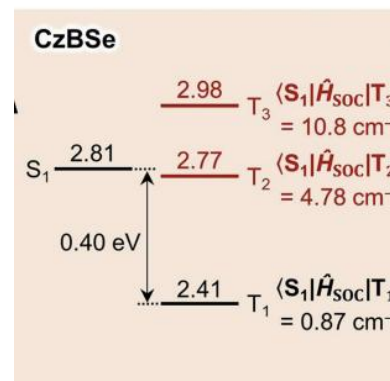
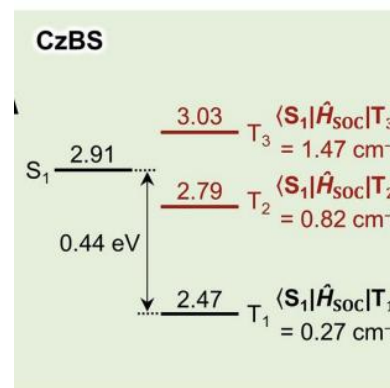
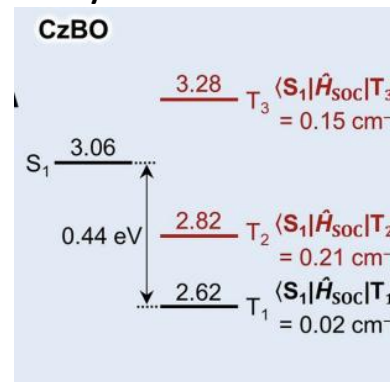


CzBSe

FWHM: 33 nm  
 $K_{\text{RISC}}/10^4 = 18000$

Conformational flexibility increased

Yasuda *et al.* Angew. Chem. Int. Ed. 2022, 61, e202205684

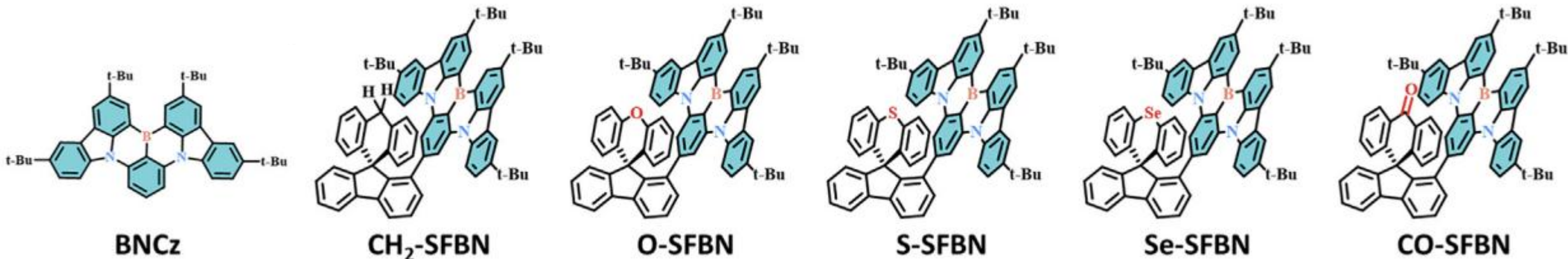


More than heavy atom effect

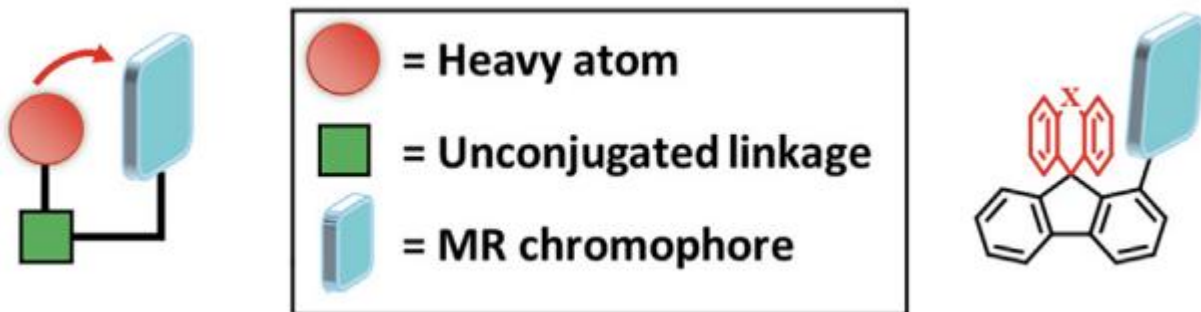
LE of Cz contribution

# Solution1: Heavy Atom Effect

Through Space Heavy atom effect

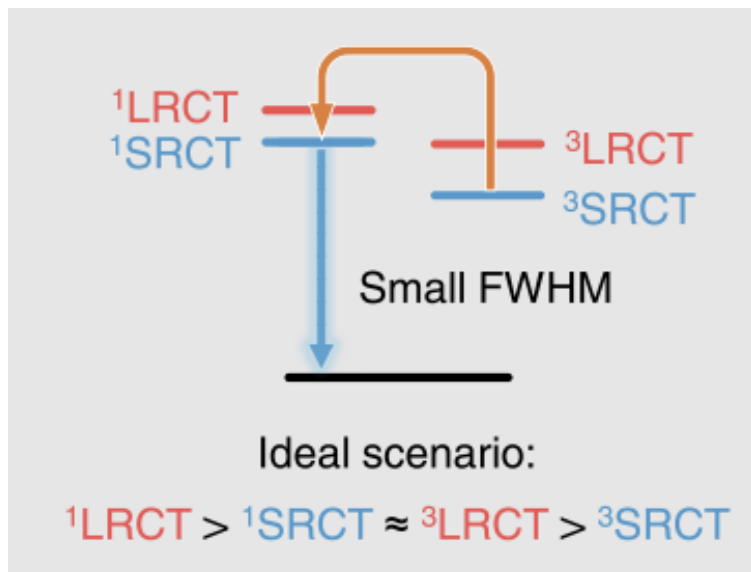


Avoid Flexibility of C-Chalo



Emitters	FWHM (nm/eV)	krISC (10 <sup>4</sup> s <sup>-1</sup> )
CH <sub>2</sub> -SFBN	25 / 0.12	0.81
O-SFBN	23 / 0.12	0.81
S-SFBN	24 / 0.12	5.16
Se-SFBN	24 / 0.12	10.50
CO-SFBN	24 / 0.12	6.64
BNCz	22 / 0.11	3.0

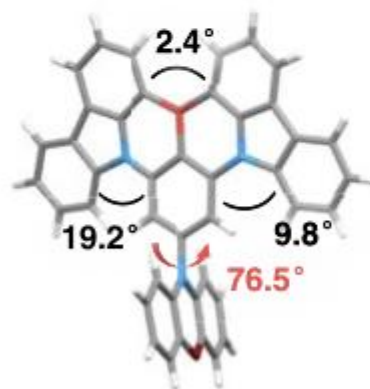
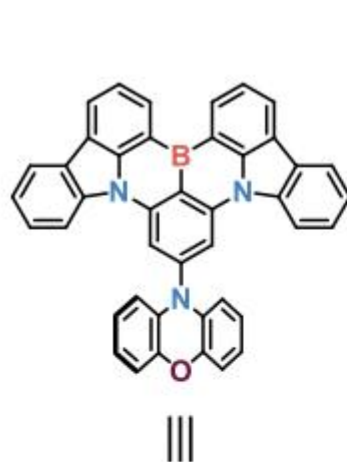
# Solution2: Transient $^3\text{LRCT}$ state



Simplified:  
 $^3\text{SRCT} \rightarrow ^3\text{LRCT} \rightarrow ^3\text{SRCT}$

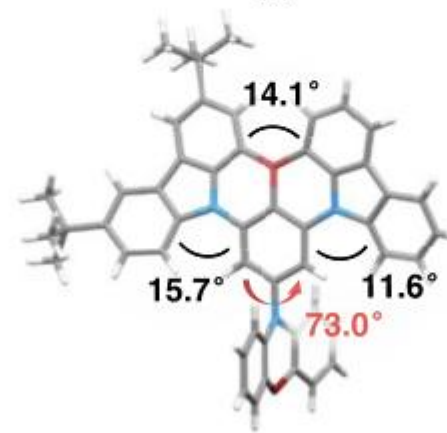
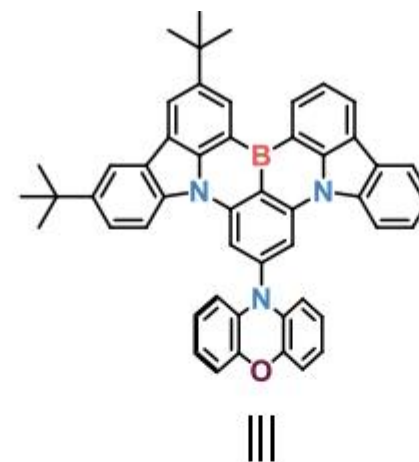
Mathematically: recalls

$$\langle S_1 | \hat{H}_{SO} | T_1 \rangle_{\text{eff}} = \frac{\langle ^1\text{CT} | \hat{H}_{SO} | ^3\text{LE} \rangle \langle ^3\text{LE} | \hat{H}_{vib} | ^3\text{CT} \rangle}{\Delta E(\text{CT} - \text{LE})}$$



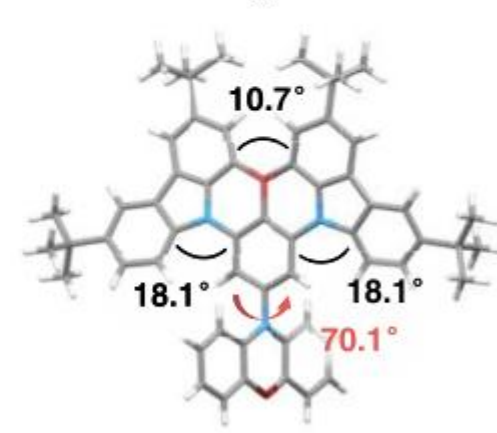
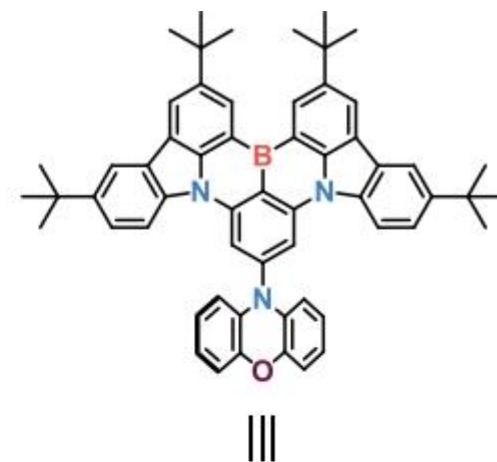
$$k_{\text{RISC}} = 2.0 \times 10^6 \text{ s}^{-1}$$

**CzBN1**



$$k_{\text{RISC}} = 7.9 \times 10^5 \text{ s}^{-1}$$

**CzBN2**



$$k_{\text{RISC}} = 3.3 \times 10^5 \text{ s}^{-1}$$

**CzBN3**

Similar  $k_{\text{RISC}}$

# Solution2: Transient $^3\text{LRCT}$ state

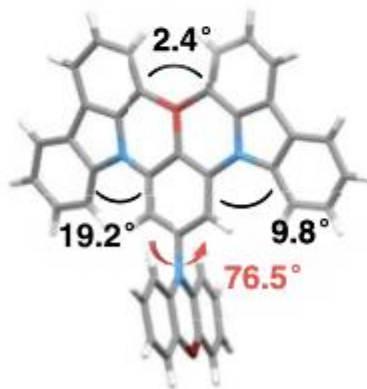
But very interestingly.....

emitter	$\lambda_{\text{abs}}^a$ [nm]	$\lambda_{\text{em}}^a$ [nm]	FWHM <sup>a</sup> [nm/eV]
BCzBN <sup>13</sup>	463	484	23/0.12
CzBN1	453	471/533	78/0.33
CzBN2	459	477/522	75/0.37
CzBN3	463	478	21/0.11

Huge difference in FWHM  
Why?

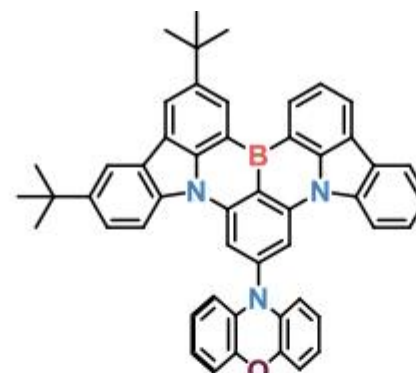


|||

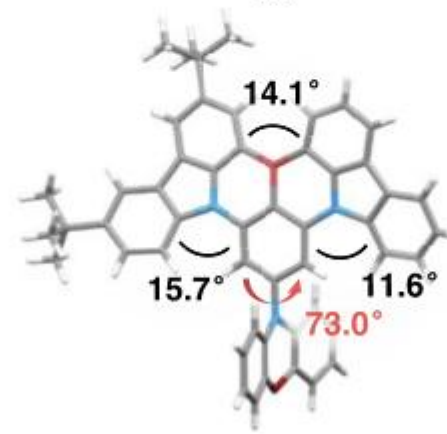


$$k_{\text{RISC}} = 2.0 \times 10^6 \text{ s}^{-1}$$

CzBN1

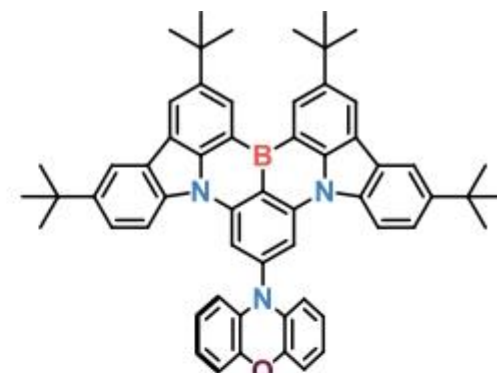


|||

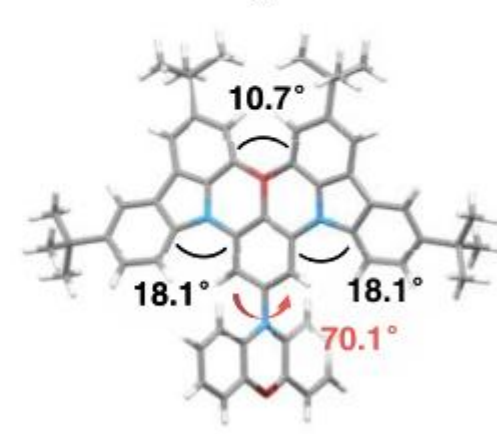


$$k_{\text{RISC}} = 7.9 \times 10^5 \text{ s}^{-1}$$

CzBN2



|||

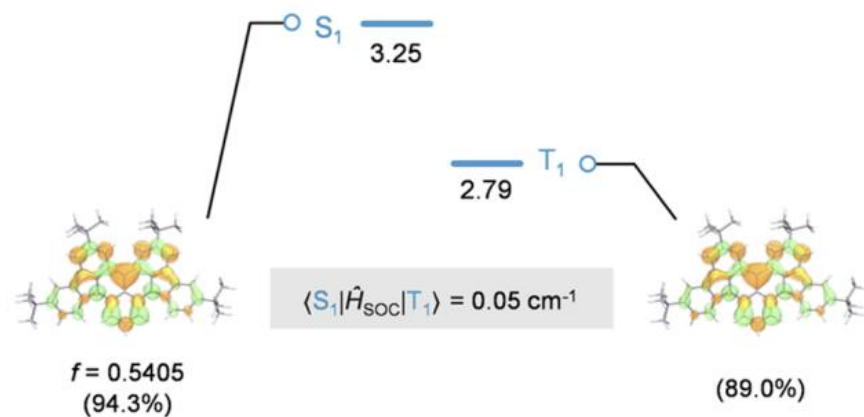


$$k_{\text{RISC}} = 3.3 \times 10^5 \text{ s}^{-1}$$

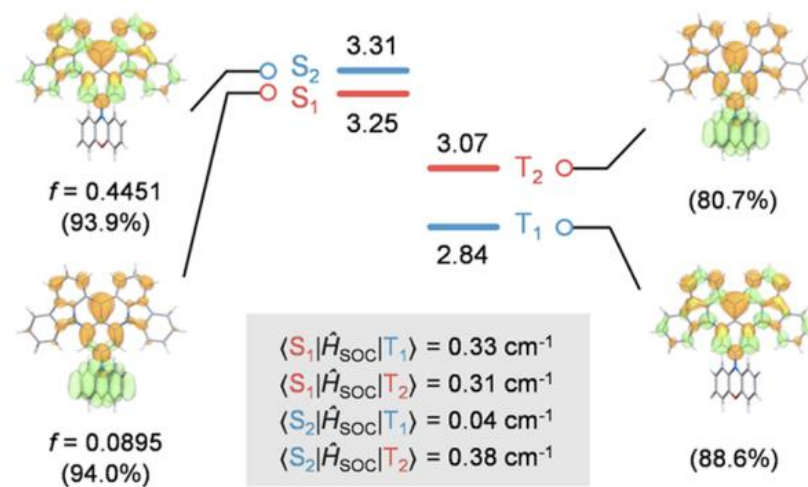
CzBN3

# Solution2: Transient $^3\text{LRCT}$ state

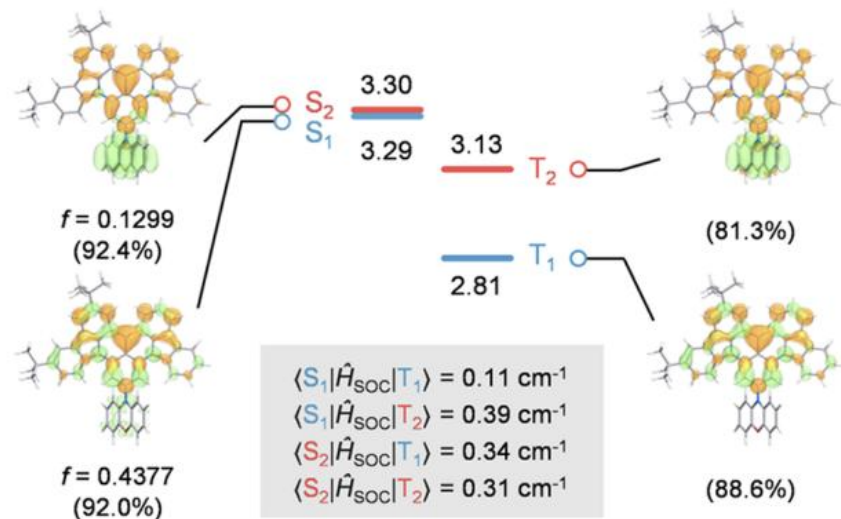
**A** BCzBN: **SRCT** emission



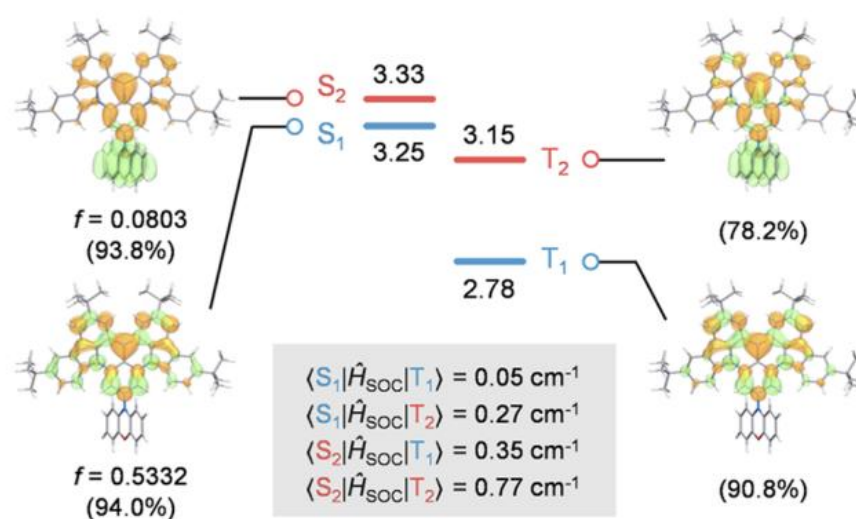
**B** CzBN1: **LRCT**-dominated emission



**C** CzBN2: **LRCT/SRCT**-hybrid emission

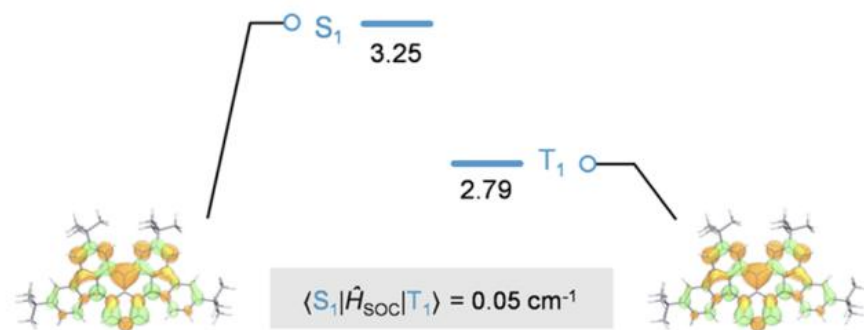


**D** CzBN3: **SRCT**-dominated emission

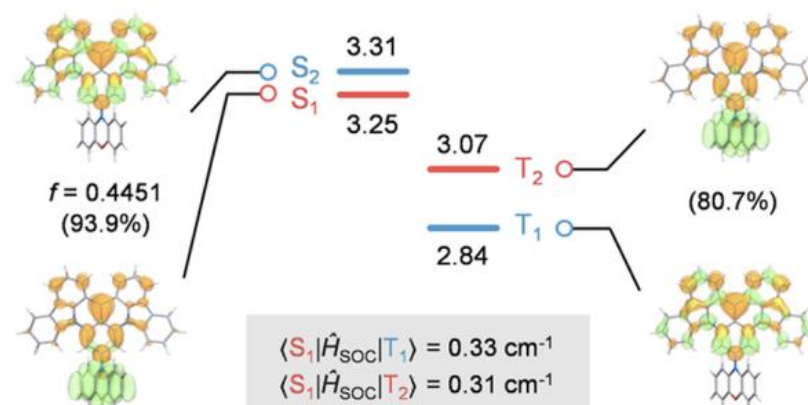


# Solution2: Transient $^3\text{LRCT}$ state

**A** BCzBN: **SRCT** emission

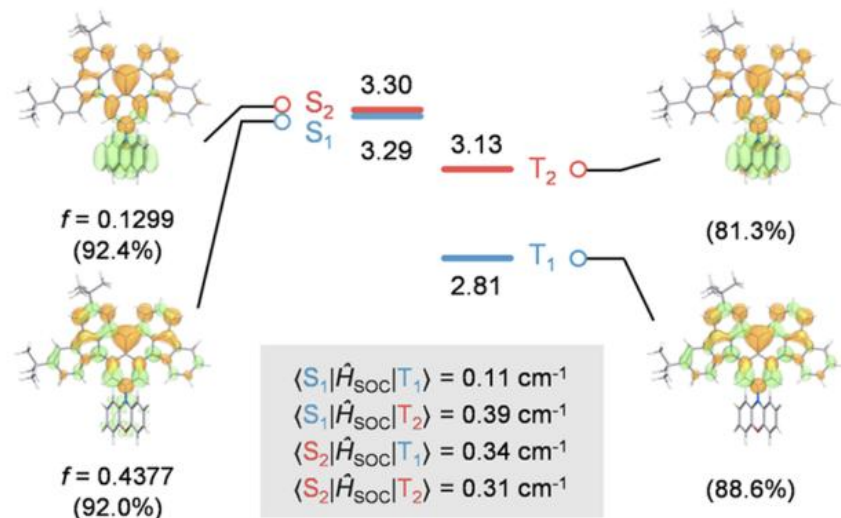


**B** CzBN1: **LRCT**-dominated emission

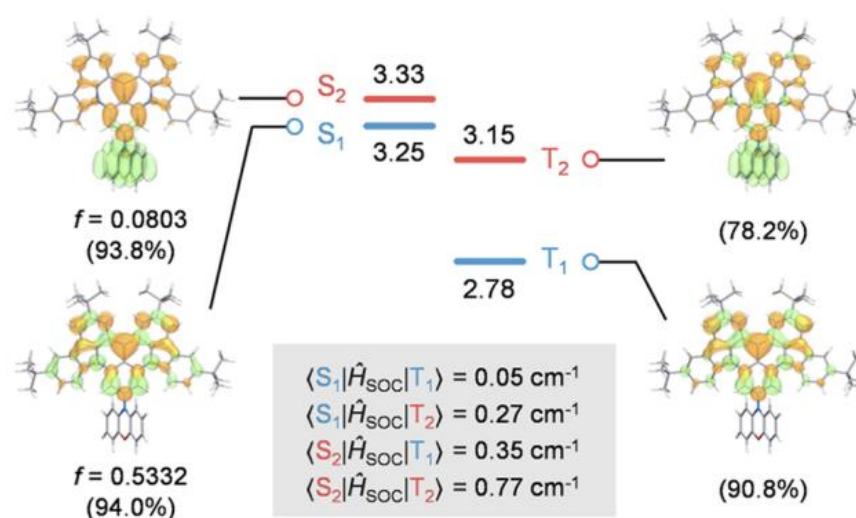


The emission should happen in  $^1\text{SRCT}$   
 $^3\text{SRCT} \gg ^1\text{SRCT} = ^3\text{LRCT} > ^3\text{SRCT}$

**C** CzBN2: **LRCT/SRCT**-hybrid emission

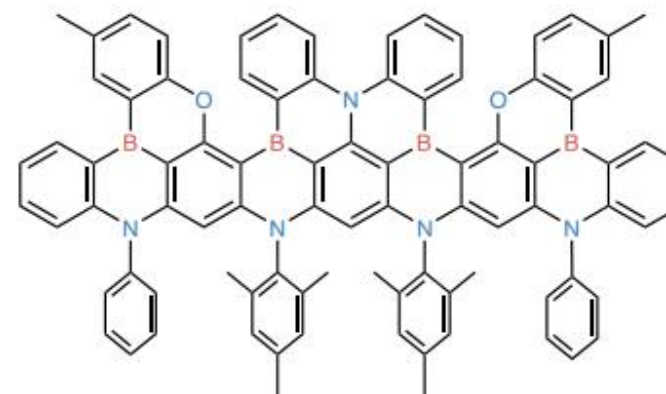
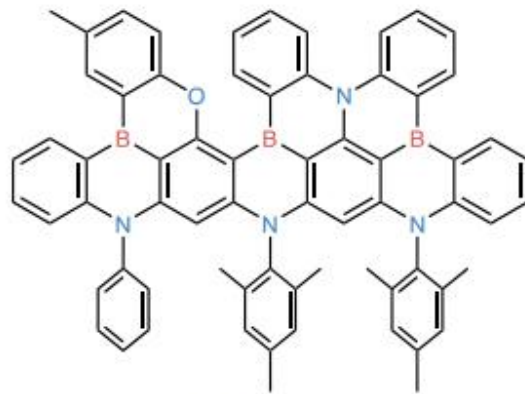
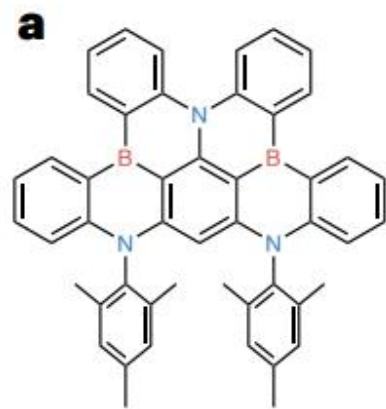


**D** CzBN3: **SRCT**-dominated emission

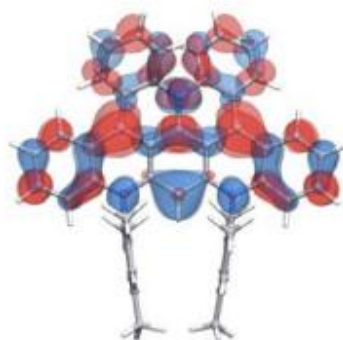


# Solution2: Transient $^3\text{LRCT}$ state

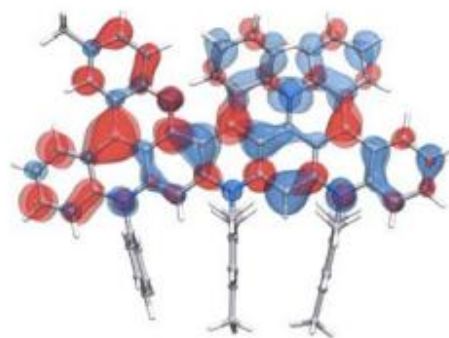
- highly twisted core
- Linear  $\pi$ -conjugation extension
- Substitution with Oxygen



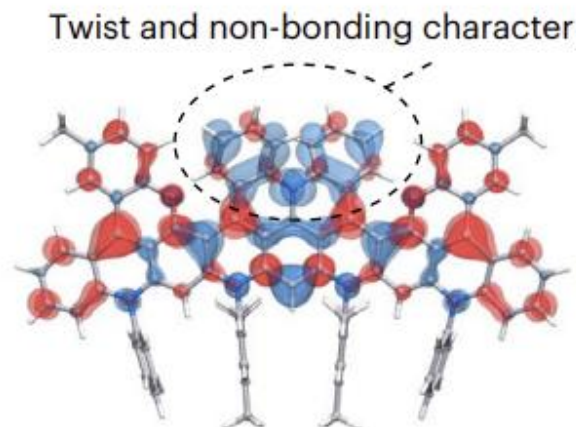
**b** ● HOMO ● LUMO



$E_g = 3.66 \text{ eV}$



$E_g = 3.43 \text{ eV}$



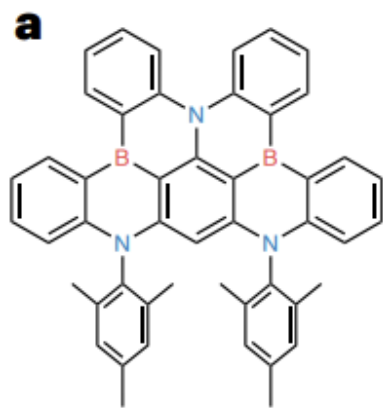
$E_g = 3.34 \text{ eV}$

# Solution2: Transient $^3\text{LRCT}$ state

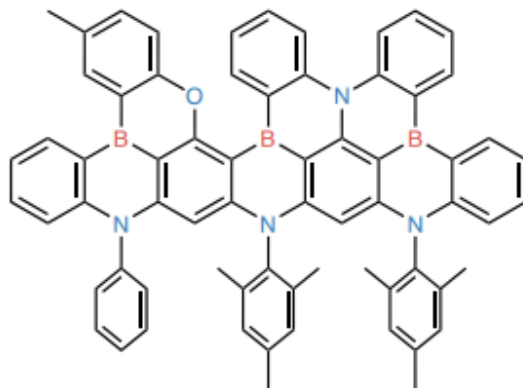
- highly twisted core

- Linear  $\pi$ -conjugation extension

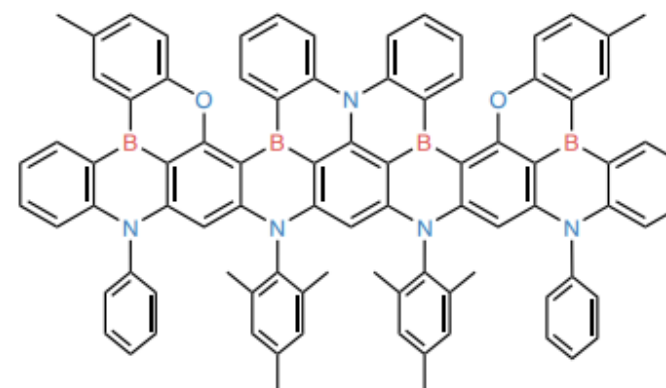
- Substitution with Oxygen



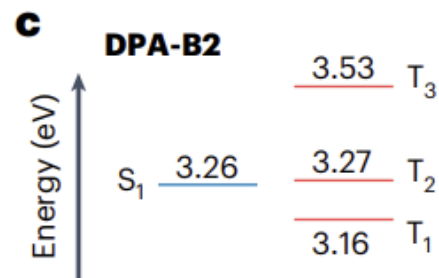
DPA-B2



DPA-B3



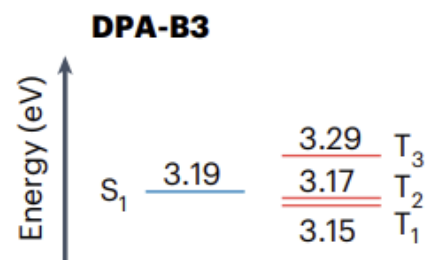
DPA-B4



$$\langle S_1 | \hat{H}_{\text{SOC}} | T_3 \rangle = 0.28 \text{ cm}^{-1}$$

$$\langle S_1 | \hat{H}_{\text{SOC}} | T_2 \rangle = 1.12 \text{ cm}^{-1}$$

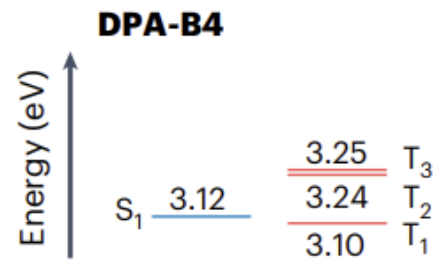
$$\langle S_1 | \hat{H}_{\text{SOC}} | T_1 \rangle = 0.15 \text{ cm}^{-1}$$



$$\langle S_1 | \hat{H}_{\text{SOC}} | T_3 \rangle = 0.28 \text{ cm}^{-1}$$

$$\langle S_1 | \hat{H}_{\text{SOC}} | T_2 \rangle = 0.88 \text{ cm}^{-1}$$

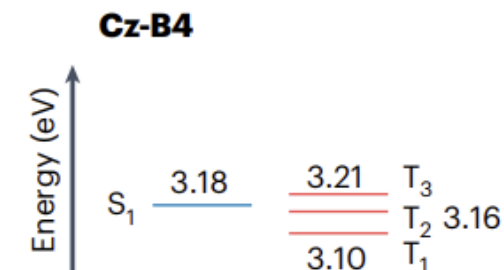
$$\langle S_1 | \hat{H}_{\text{SOC}} | T_1 \rangle = 0.26 \text{ cm}^{-1}$$



$$\langle S_1 | \hat{H}_{\text{SOC}} | T_3 \rangle = 0.66 \text{ cm}^{-1}$$

$$\langle S_1 | \hat{H}_{\text{SOC}} | T_2 \rangle = 0.63 \text{ cm}^{-1}$$

$$\langle S_1 | \hat{H}_{\text{SOC}} | T_1 \rangle = 0.02 \text{ cm}^{-1}$$



$$\langle S_1 | \hat{H}_{\text{SOC}} | T_3 \rangle = 0.08 \text{ cm}^{-1}$$

$$\langle S_1 | \hat{H}_{\text{SOC}} | T_2 \rangle = 0.61 \text{ cm}^{-1}$$

$$\langle S_1 | \hat{H}_{\text{SOC}} | T_1 \rangle = 0.05 \text{ cm}^{-1}$$

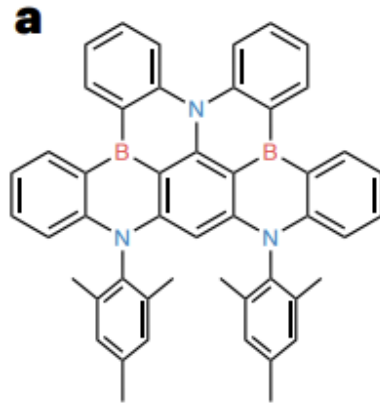
Ultra-fast RISC Rate ( $2.29 \times 10^6 \text{ s}^{-1}$ ) without Heavy Atoms

# Solution2: Transient $^3\text{LRCT}$ state

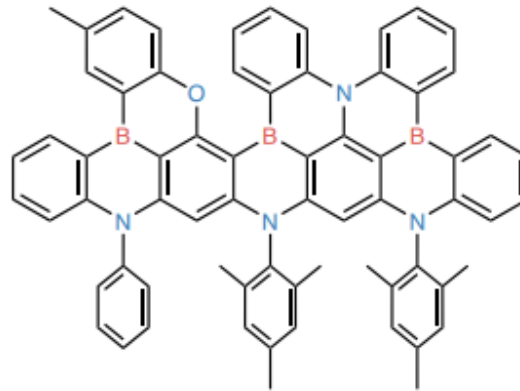
- highly twisted core

- Linear  $\pi$ -conjugation extension

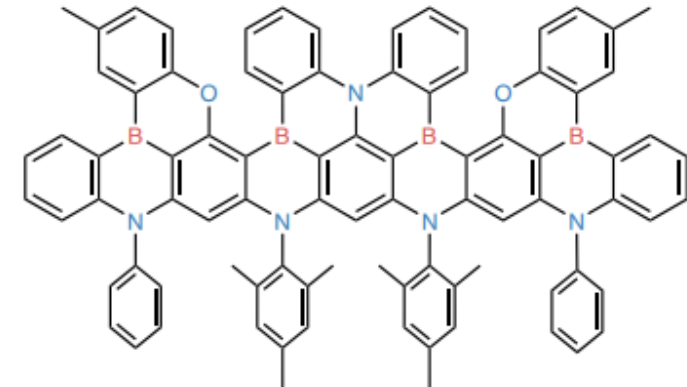
- Substitution with Oxygen



DPA-B2



DPA-B3



DPA-B4

Emitter	$\lambda_{\text{abs}}^{\text{a}}$ (nm)	$\lambda_{\text{em}}^{\text{a}}$ (nm)	Ultra Sharp
			FWHM <sup>a</sup> (nm)
DPA-B2	424	440 (444)	27 (31)
DPA-B3	438	448 (451)	14 (16)
DPA-B4	448	458 (458)	12 (14)

Deep Blue

EQE=39.2%(single device)  
/74.5%(hyperfluorescence device)

Current State of the art

# ***MR-TADF*s**

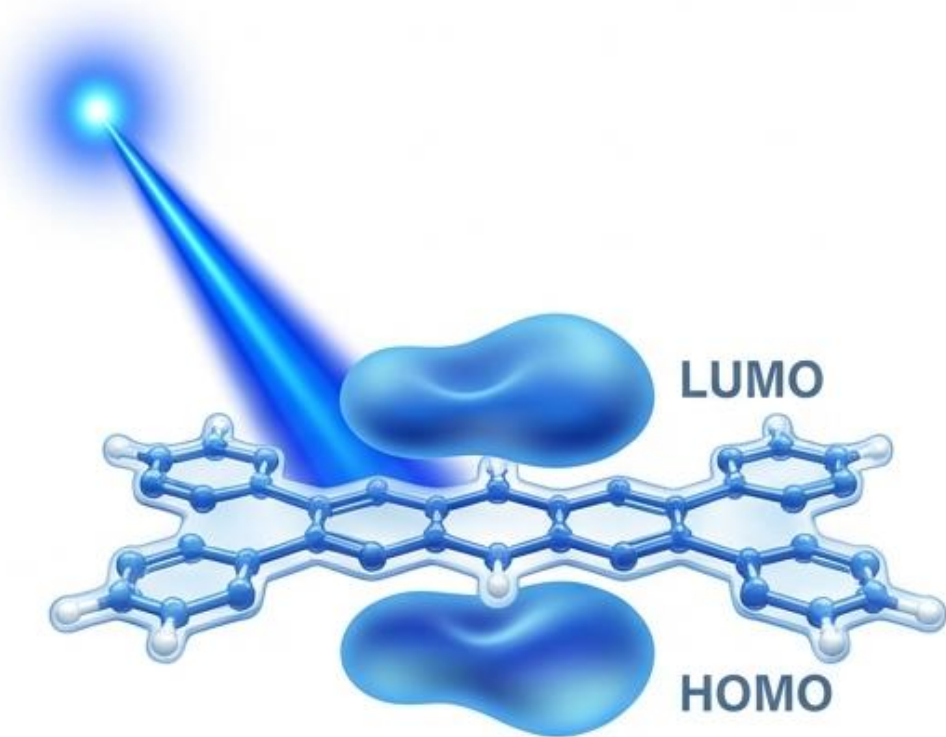
## ***Content***

- *TADFs and Essential Theoretical Backgrounds*
- *D-A type TADFs and their limitations*
- *MR-TADFs*
- *Towards Deep RED Emission*
- *Towards Brighter OLEDs: faster RISC*
- *Towards Brighter OLEDs: against ACQs*

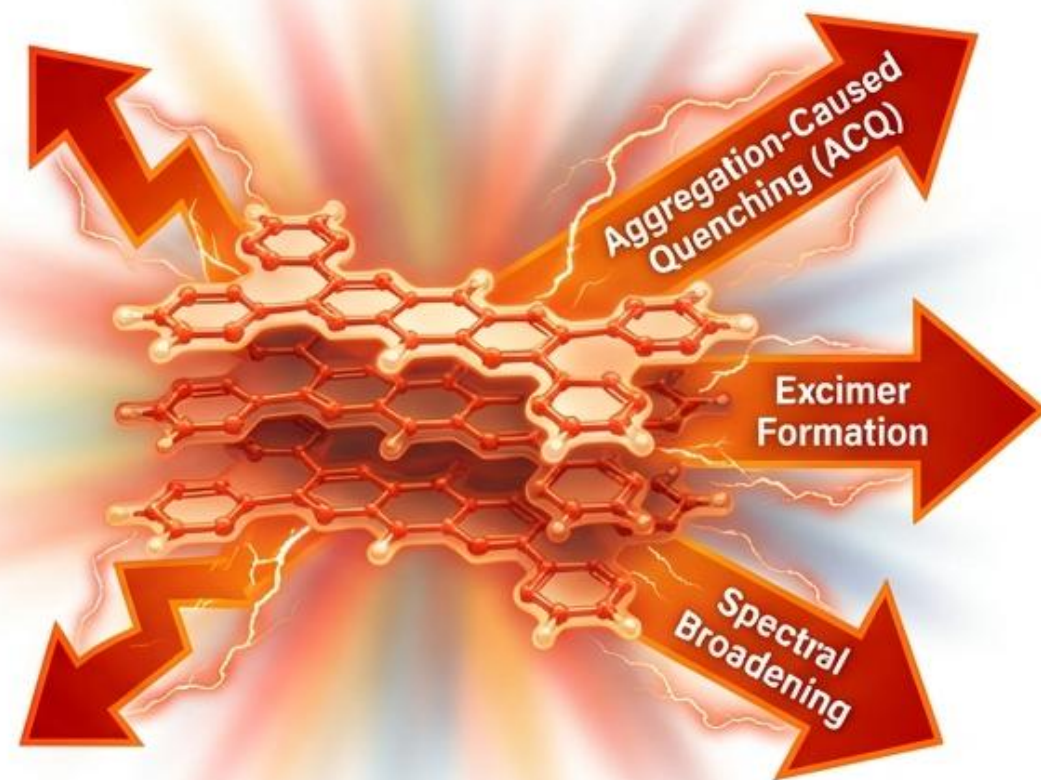
The Ideal

# The Planar Perfection of MR-TADF Cores is Their Primary Downfall

The Reality

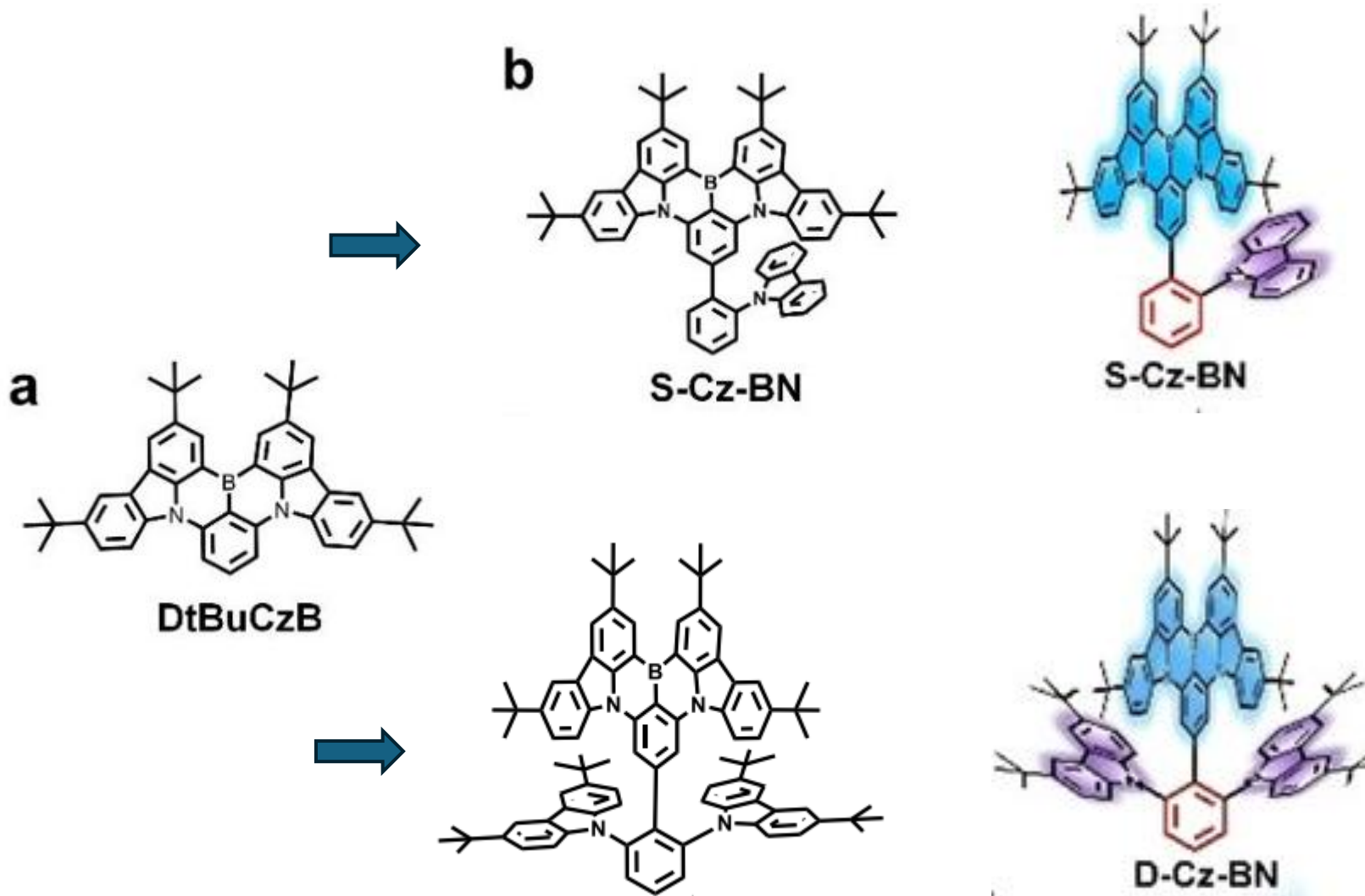


Rigid, planar polycyclic frameworks enable pristine narrowband emission, but cause fatal interchromophore  $\pi$ - $\pi$  stacking at high doping ratios.



The result is broadened spectra, quenched external quantum efficiency (EQE), and severe triplet annihilation.

# Introduction of steric hindrance

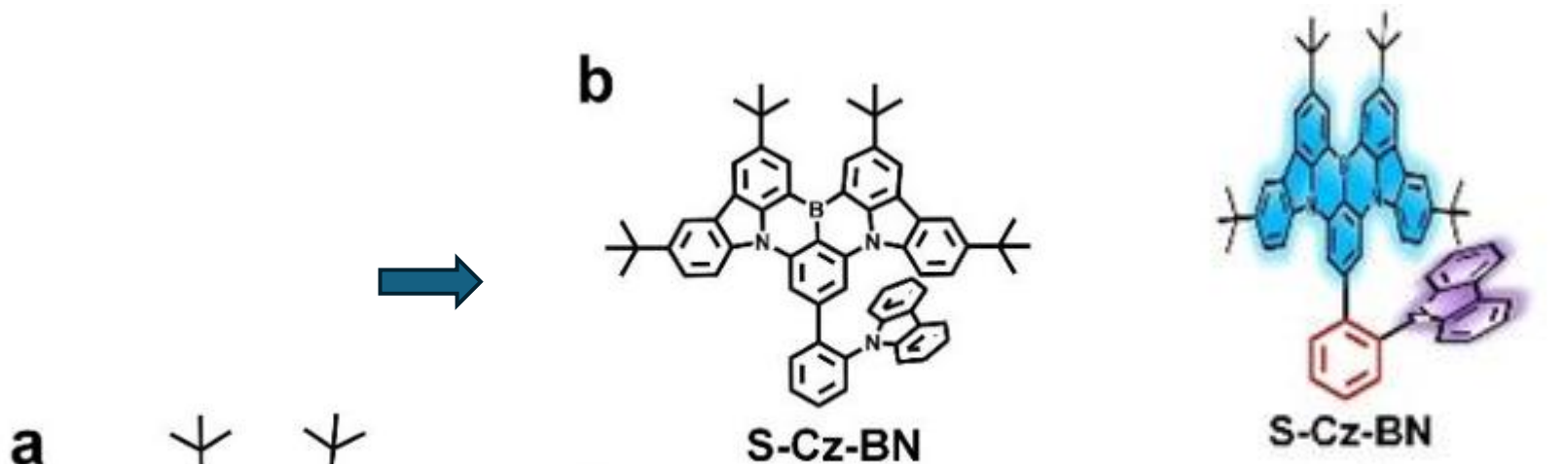


	FWHM (nm)	kRISC (10 <sup>4</sup> s <sup>-1</sup> )
<b>DtBuCzB</b>	25	3.0
<b>S-Cz-BN</b>	23	1.8
<b>D-Cz-BN</b>	22	1.8

**Table 2:** Device performances of the MR-TADF based C

Devices	$\lambda_{EL}^{[a]}$ [nm]	FWHM <sup>[b]</sup> [nm]	EQE <sup>[d]</sup> [%]
1 wt% DtBuCzB	488	29	27.5/26.9/2
20 wt% DtBuCzB	496	37	18.1/17.3/1
1 wt% S-Cz-BN	488	26	30.5/30.2/2
20 wt% S-Cz-BN	494	31	28.8/26.9/1
1 wt% D-Cz-BN	488	24	37.2/37.2/3
20 wt% D-Cz-BN	491	24	36.3/36.2/2

# Introduction of steric hindrance



	FWHM (nm)	kRISC (10 <sup>4</sup> s <sup>-1</sup> )
DtBuCzB	25	3.0
S-Cz-BN	23	1.8
D-Cz-BN	22	1.8

NOT Current state of the art  
But non of the following work is interesting enough to discuss here

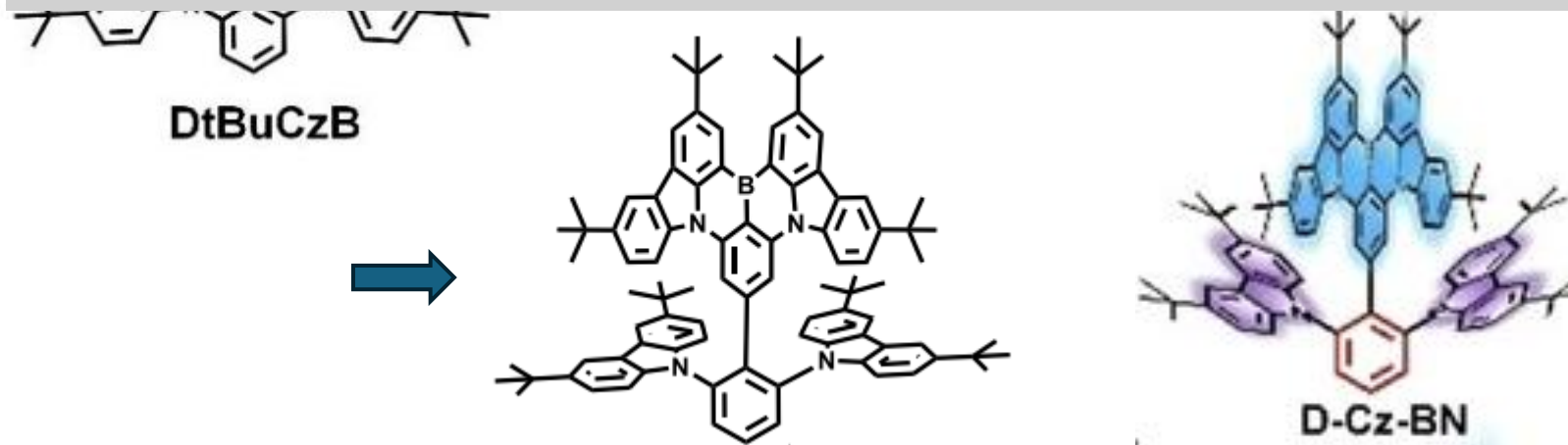


Table 2: Device performances of the IRK-TADF based C

Devices	$\lambda_{EL}^{[a]}$ [nm]	FWHM <sup>[b]</sup> [nm]	EQE <sup>[d]</sup> [%]
1 wt% DtBuCzB	488	29	27.5/26.9/2
20 wt% DtBuCzB	496	37	18.1/17.3/1
1 wt% S-Cz-BN	488	26	30.5/30.2/2
20 wt% S-Cz-BN	494	31	28.8/26.9/1
1 wt% D-Cz-BN	488	24	37.2/37.2/3
20 wt% D-Cz-BN	491	24	36.3/36.2/2

# Conclusion

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- *Balancing  $\Delta E_{ST}$ ,  $f$ , and  $H_{soc}$*
- *D-A type TADFs and their limitations: Huge FWHM*
- *MR-TADFs gives small FWHM (SRCT)*
- *Towards Deep RED Emission through controlling SRCT and bigger BN ring*
- *Faster RISC: Heavy atom effect + combining other state*
- *Against ACQs: Introducing sterics*