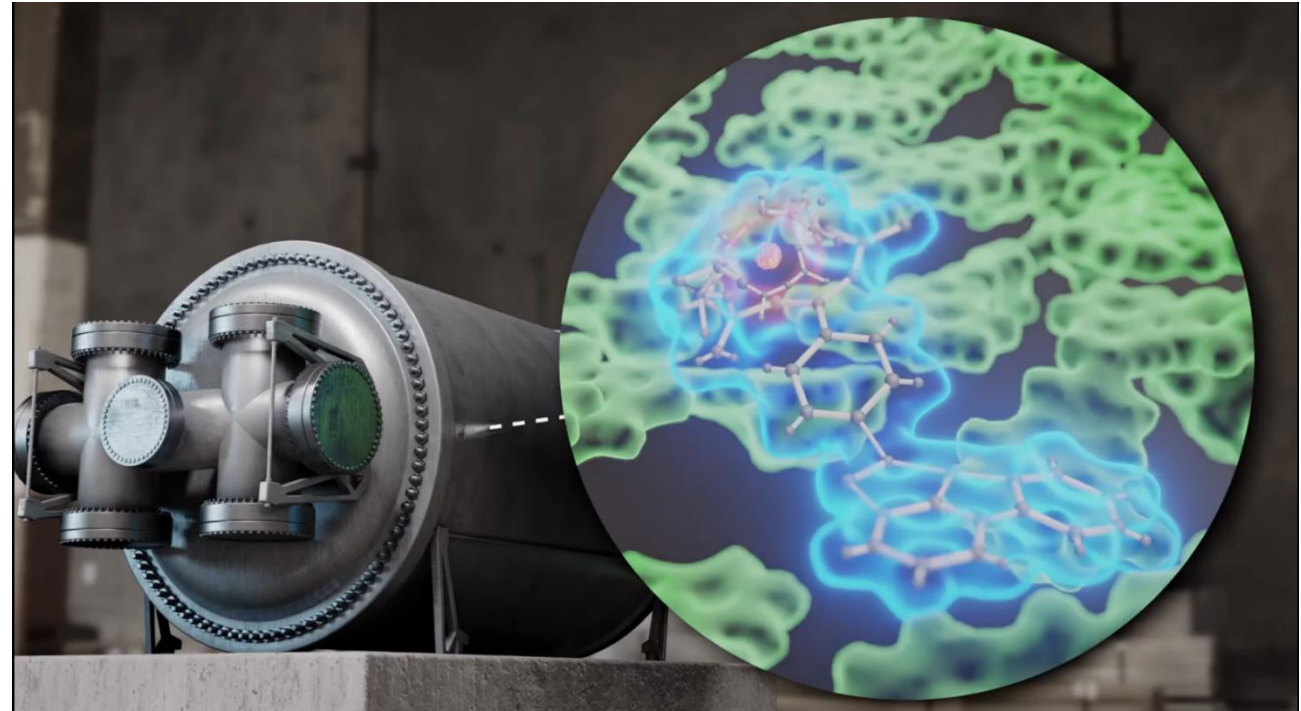

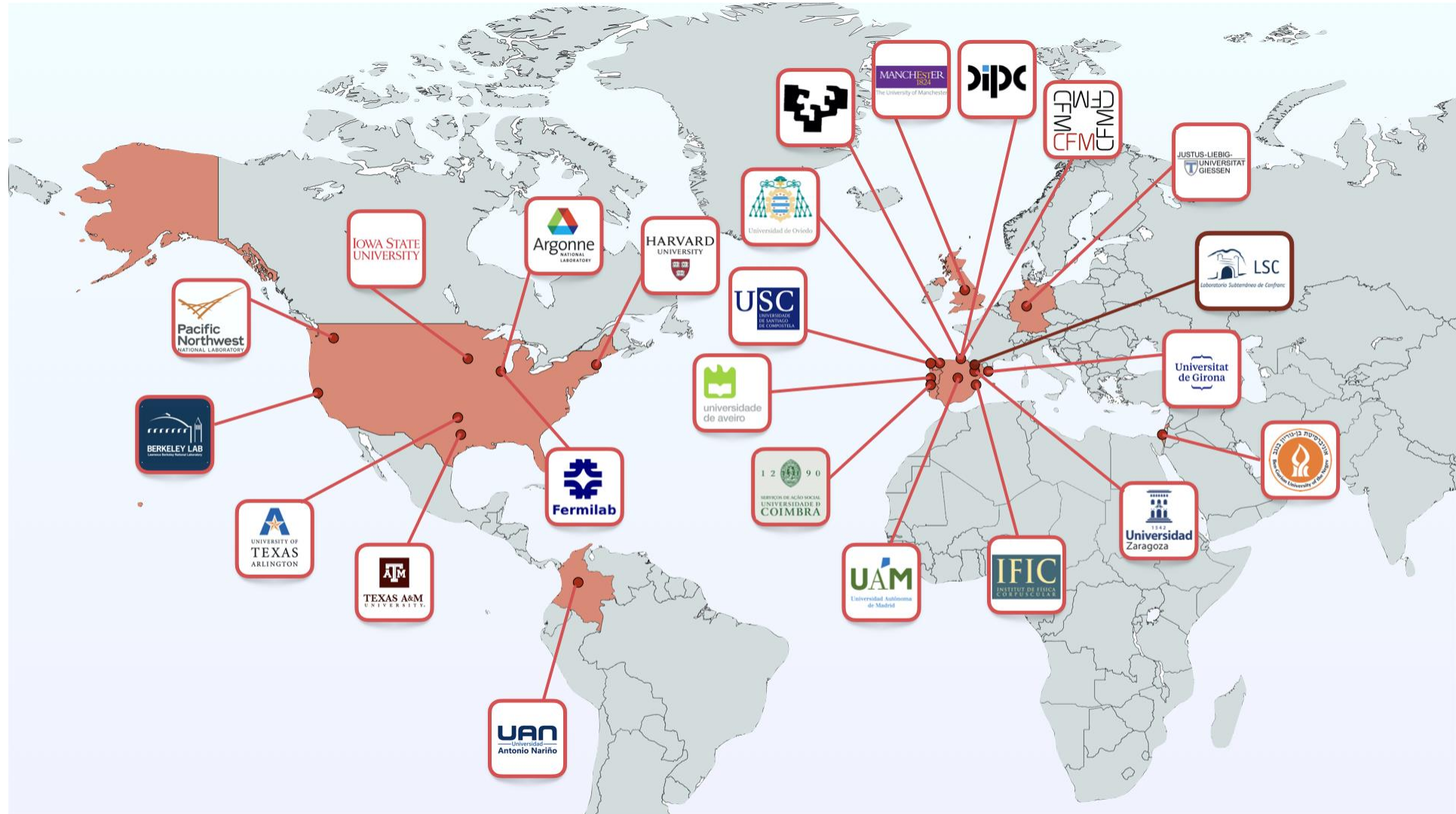


Searching the Grail: A background free $\beta\beta 0\nu$ experiment using Ba^{2+} tagging in a High-Pressure Xenon Chamber



Iván Rivilla and on behalf of 

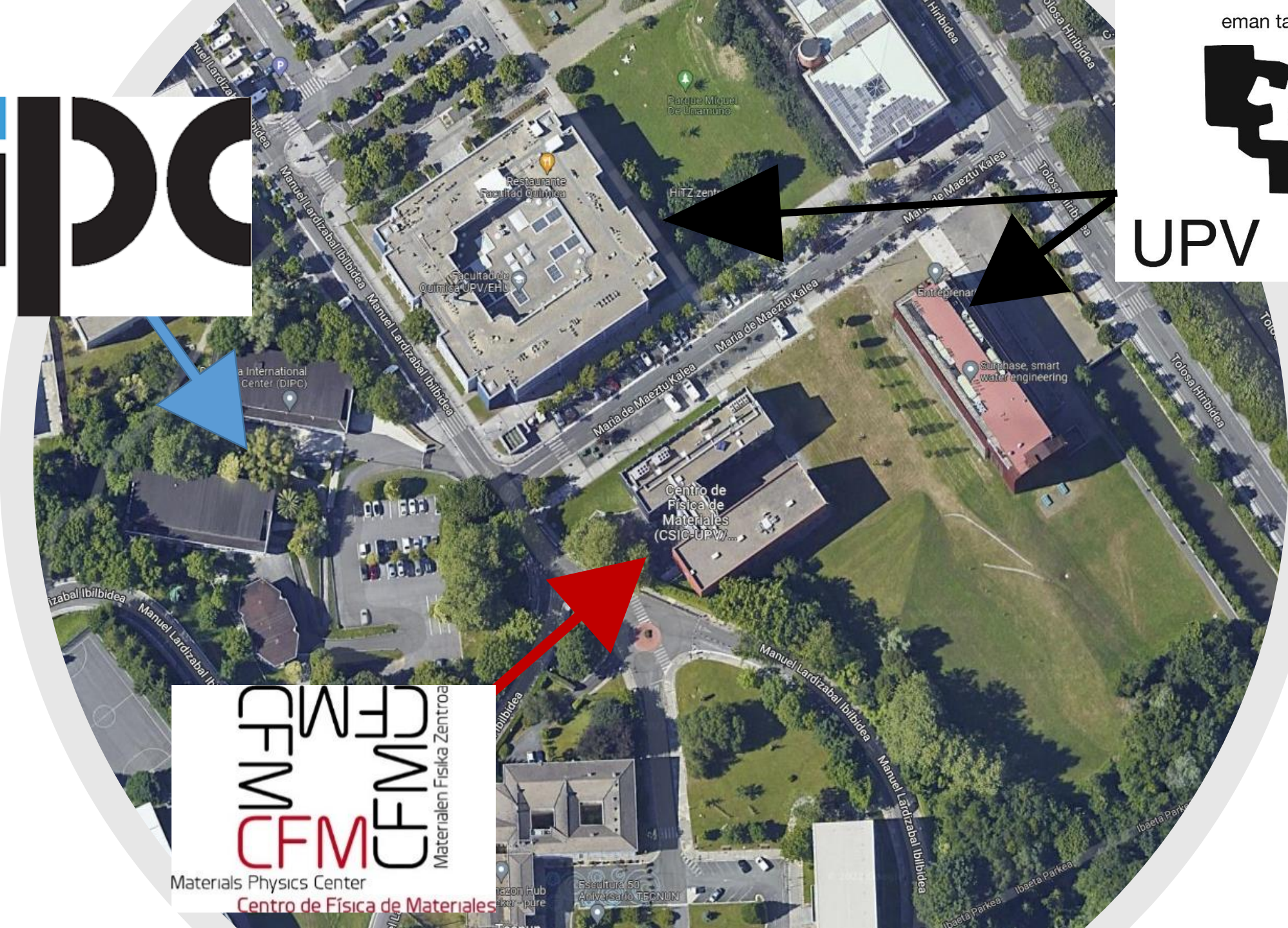




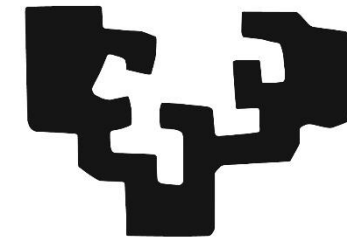




DIPC



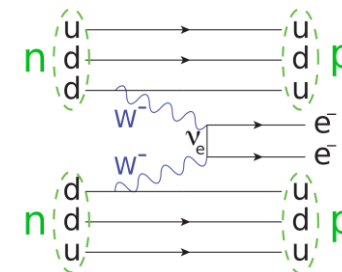
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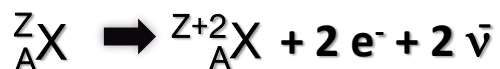
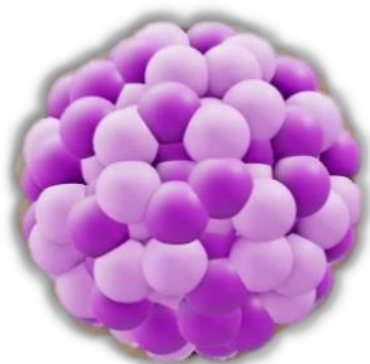
UPV EHU

CFM CFM CFM
Materials Physics Center
Centro de Física de Materiales

How to confirm experimentally if neutrino is a Majorana particle?



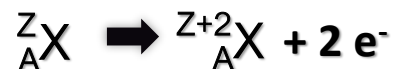
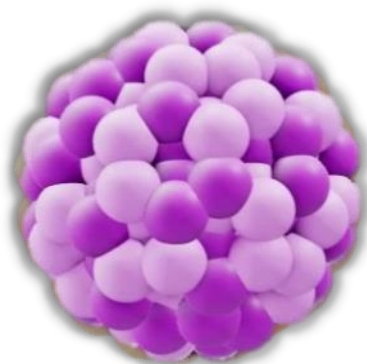
$\beta\beta 2\nu$



SM-allowed process
Measured in several nuclei

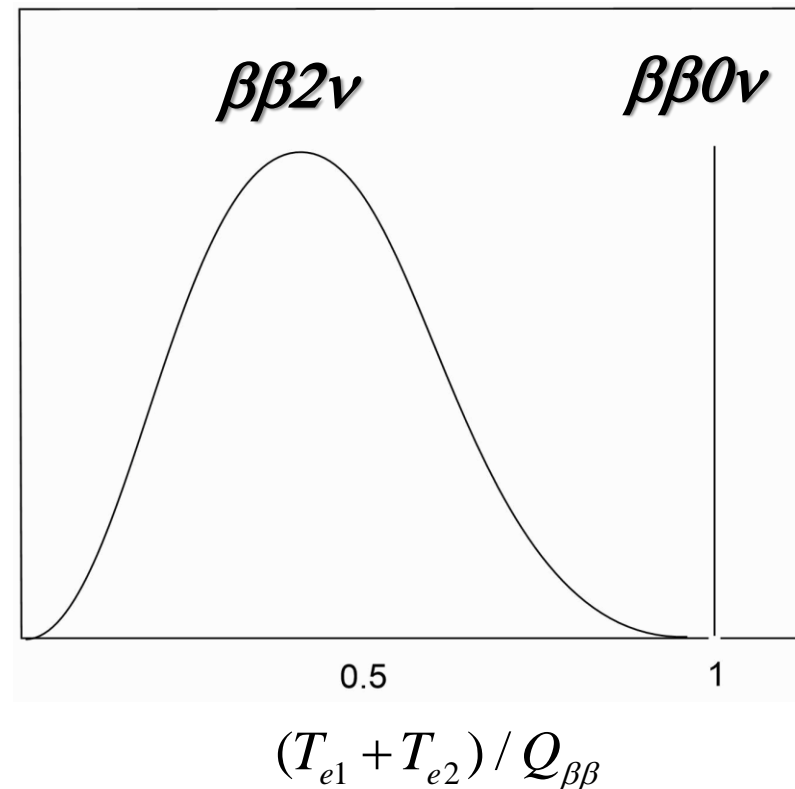
$$T_{1/2}^{2\nu} \sim 10^{19} - 10^{21} \text{ yr}$$

$\beta\beta 0\nu$

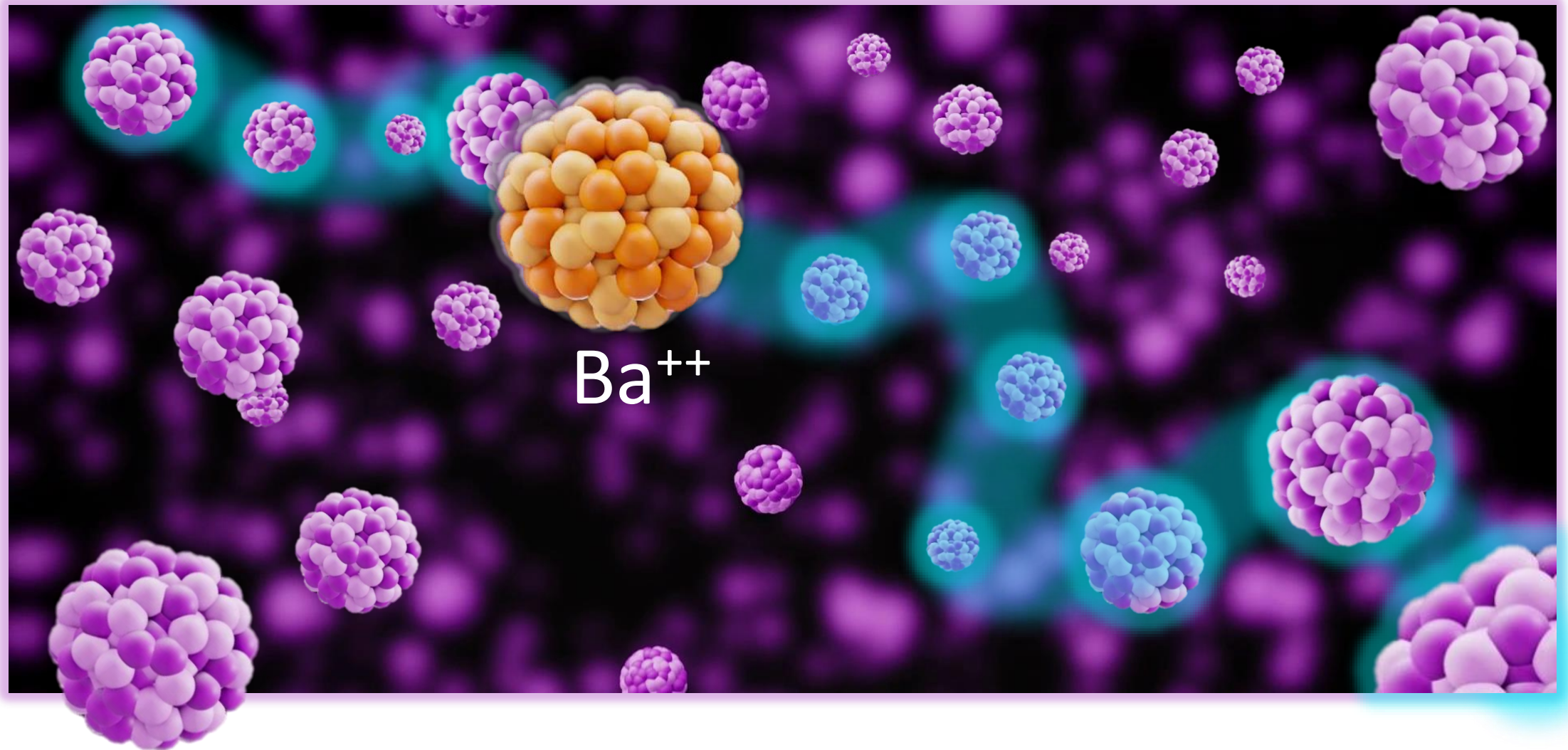


Lepton number violating process
Requires massive, Majorana neutrinos

$$T_{1/2}^{0\nu} > 10^{27} \text{ yr}$$



How to confirm experimentally if neutrino is a Majorana particle?



Detecting “tagging” the Ba⁺⁺ signaling a $\beta\beta 0\nu$ process has been a long sought holy grail of xenon chambers.

A. Usón
Talk

The NEXT Program

Prototypes
2010-2014
Demonstration of detector concept

~1 kg

NEXT-White
2015-2021
Background model assessment
 $2\nu\beta\beta$ measurement for ^{136}Xe

~5 kg

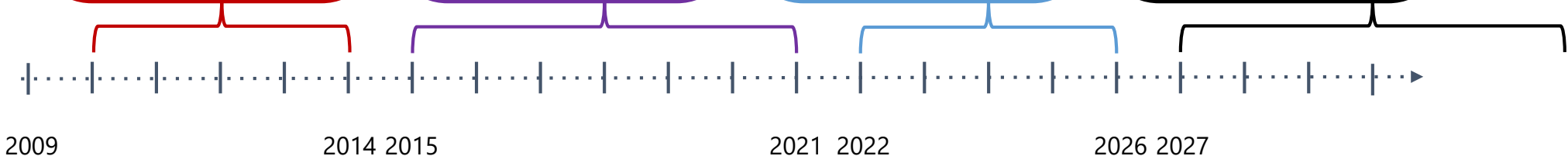
NEXT-100
2022-2026
Neutrinoless double beta decay search in ^{136}Xe

~100 kg

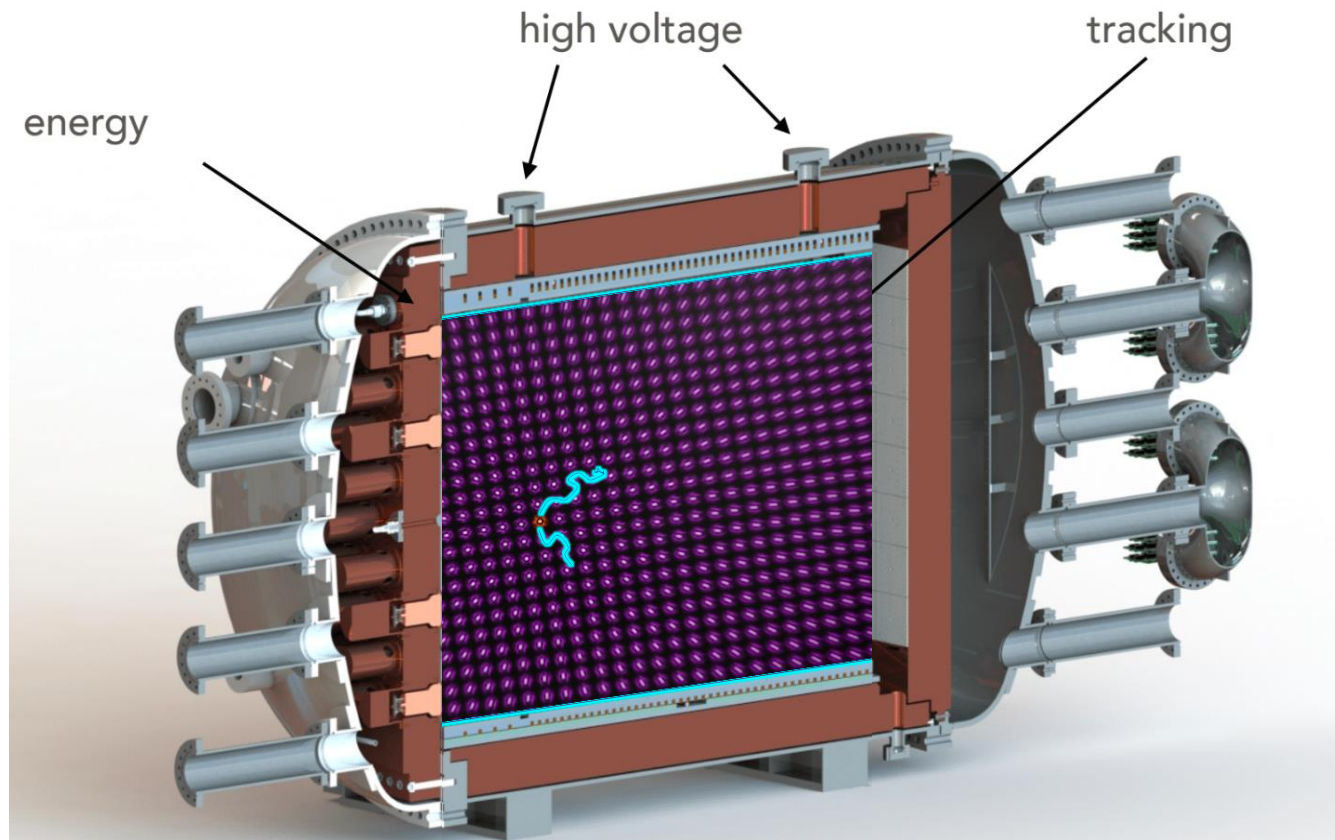
NEXT-HD
2027?
Neutrinoless double beta decay search through inverted neutrino mass ordering

NEXT-BOLD
Barium tagging for background-free experiment

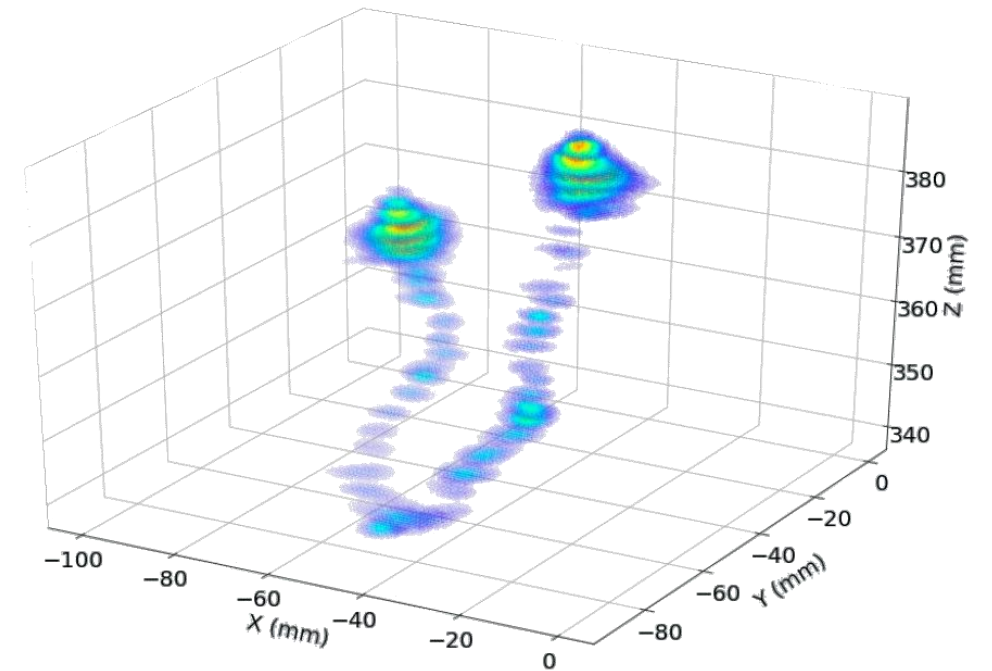
1 Tonne



NEXT detectors capabilities



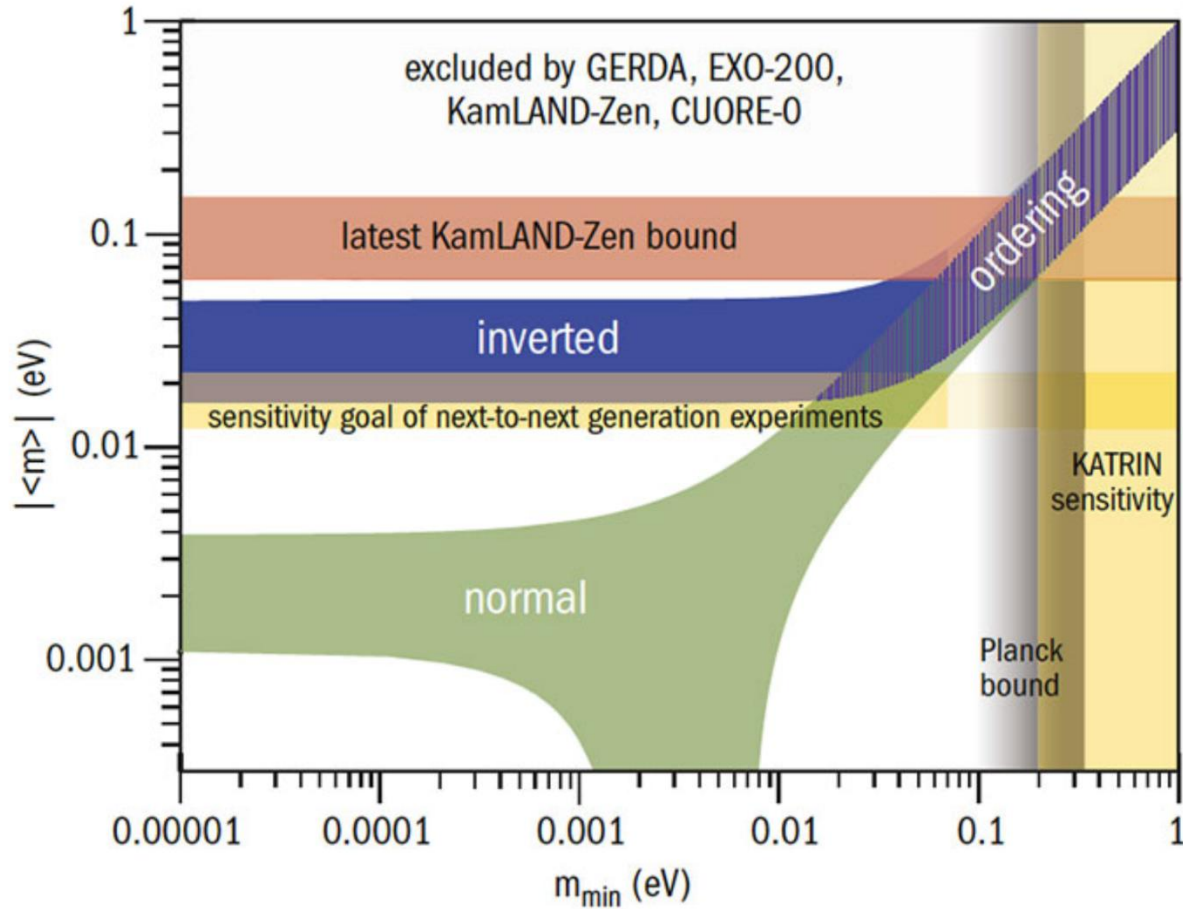
$\beta\beta 0\nu$ in high-pressure ^{136}Xe gas



$\beta\beta 2\nu$ candidate at 2 MeV, showing 2 energy blobs at the extremes.
Energy resolution better than 1% FWHM at $Q_{\beta\beta}$.

Detecting “tagging” the Ba^{++} signaling a $\beta\beta 0\nu$ process has been a long sought holy grail of xenon chambers.

The NEXT thing:



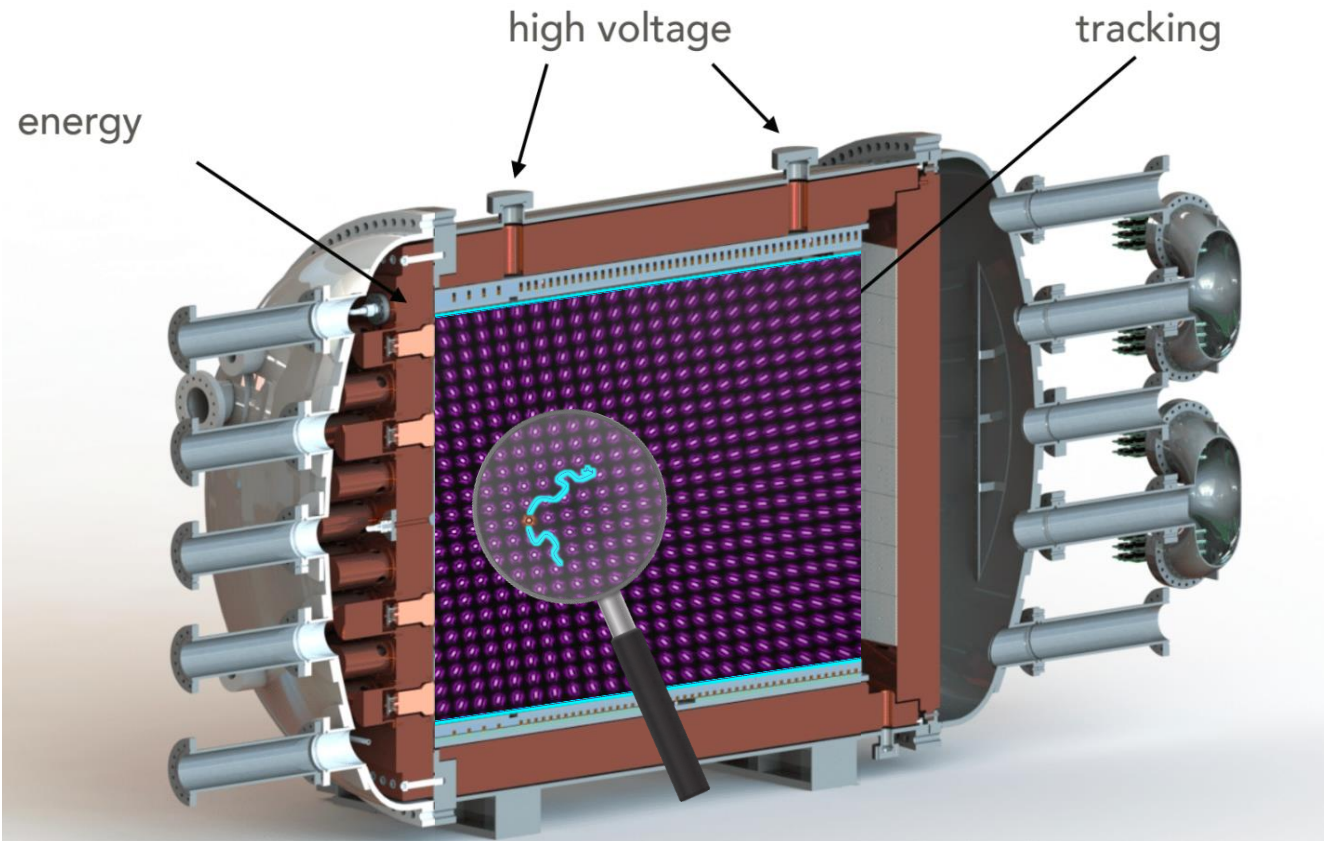
Current generation detectors “touching” inverse hierarchy región.

Objective for the next generation is to cover this region.

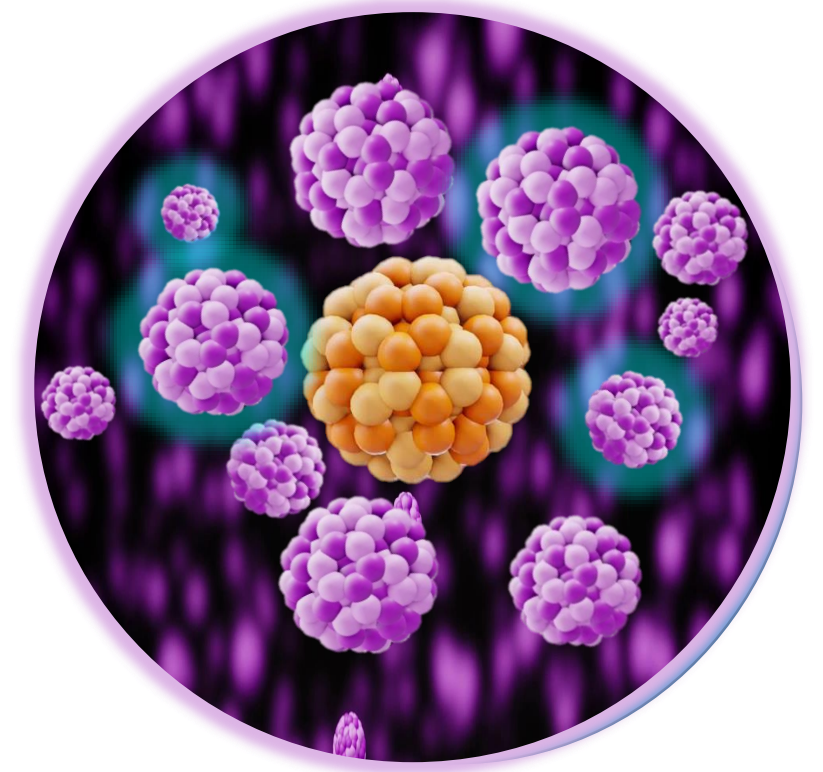
Lifetimes $\sim 10^{27-28}$ years

This implies almost background free detectors at the *ca.*Tonne-scale

How to get there with xenon gaseous detectors:



Ba⁺⁺



$\beta\beta\nu$ in high-pressure ^{136}Xe gas

Detecting “tagging” the Ba⁺⁺ signaling a $\beta\beta\nu$ process has been a long sought holy grail of xenon chambers.

Detecting the barium daughter in ^{136}Xe $0\nu\beta\beta$ decay using single-molecule fluorescence imaging techniques

David R. Nygren

Department of Physics, University of Texas at Arlington
Box 19059, Arlington, TX 76015

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Abstract. Single-molecule fluorescent imaging may provide an avenue to efficiently detect the Ba^{++} daughter atom in the decay $^{136}\text{Xe} \rightarrow \text{Ba} + 2e^-$, and, unambiguously associate the birth point in space within the electron trajectories of the decay event. Chelation of doubly-charged alkaline earth elements such as calcium and barium by certain precursor molecules converts the resulting complex from a non-fluorescent to a fluorescent state. Repeated photo-excitation of a single fluorescent complex reveals both presence and location with high precision. This technique, widespread now in biochemistry, biophysics and biology, may permit a similar discriminating response in a large high-pressure xenon gas TPC for the Ba^{++} ion from xenon double-beta decay. The TPC measures the event time and energy of the two nascent electrons, as well as topology and position in 3-D from their trajectories in the gas. Measurement of the 2-D location of the molecular ion after arrival at the cathode plane permits an association of ion with the event. Demonstration of an efficient, highly specific detection of the barium daughter would provide a long-sought pathway to a background-free result in the search for this decay mode, of central importance for determining the nature of the neutrino.

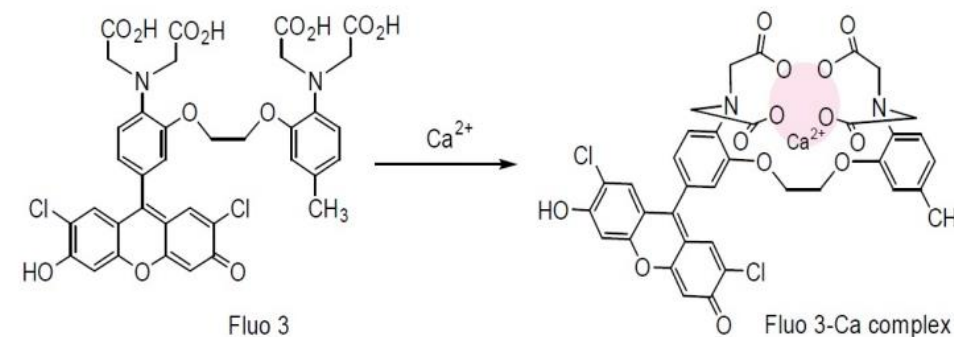
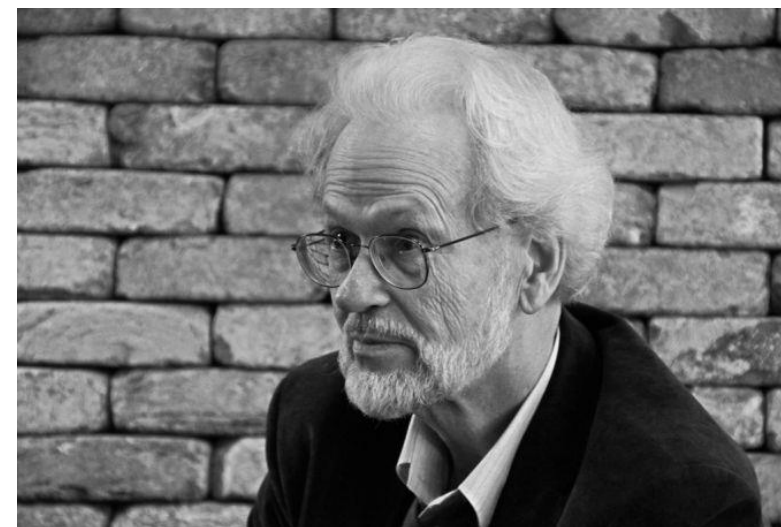


Figure 1. The skeletal formula for Fluo-3 is shown before and after complex formation. The conformational change that Fluo-3 undergoes in chelation with Ca^{++} creates a fluorescent complex. In that state, the fluorescent response increases by a factor of 60 - >100 in the cellular environment. *Image source:* Dojindo Molecular Industries, Inc.

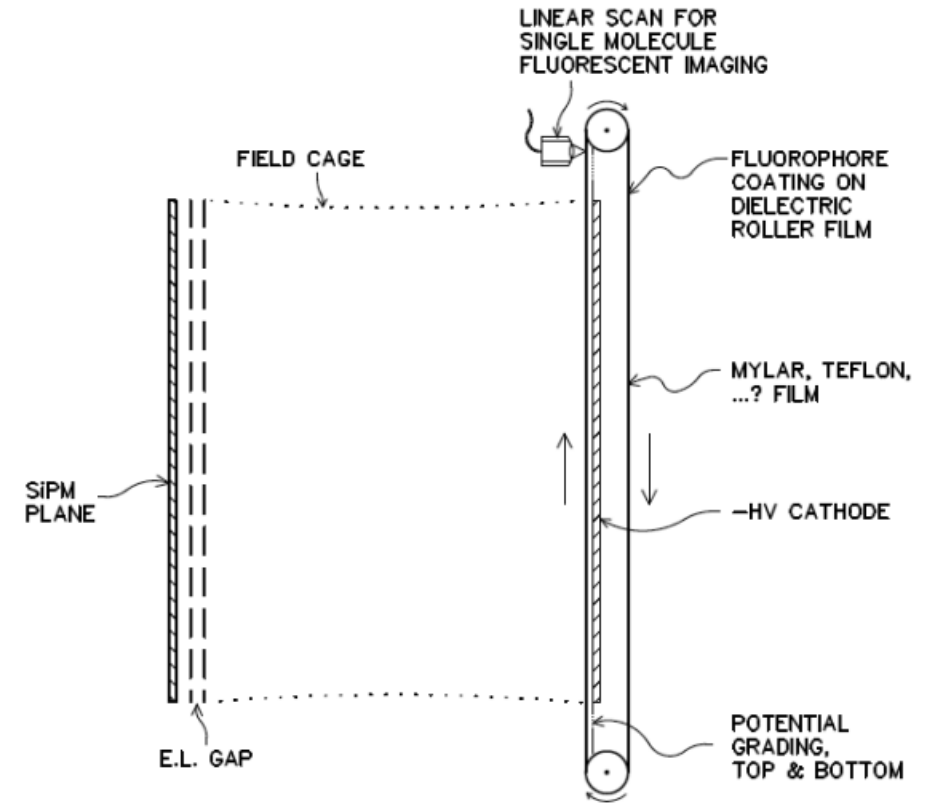
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HIGH PRESSURE XENON GAS ELECTROLUMINESCENT TPC WITH SINGLE MOLECULE FLUORESCENT IMAGING OF BARIUM DAUGHTER

Demonstration of Single-Barium-Ion Sensitivity for Neutrinoless Double-Beta Decay Using Single-Molecule Fluorescence Imaging

A. D. McDonald,^{1,†} B. J. P. Jones,^{1,‡} D. R. Nygren,^{1,§} C. Adams,² V. Álvarez,³ C. D. R. Azevedo,⁴ J. M. Benlloch-Rodríguez,³ F. I. G. M. Borges,⁵ A. Botas,³ S. Cárcel,³ J. V. Carrión,³ S. Cebrián,⁶ C. A. N. Conde,⁵ J. Díaz,³ M. Diesburg,⁷ J. Escada,⁵ R. Esteve,⁸ R. Felkai,³ L. M. P. Fernandes,⁹ P. Ferrario,³ A. L. Ferreira,⁴ E. D. C. Freitas,⁹ A. Goldschmidt,¹⁰ J. J. Gómez-Cadenas,^{3,§} D. González-Díaz,¹¹ R. M. Gutiérrez,¹² R. Guenette,² K. Hafidi,¹³ J. Hauptman,¹⁴ C. A. O. Henriques,⁹ A. I. Hernandez,¹² J. A. Hernando Morata,¹¹ V. Herrero,⁸ S. Johnston,¹⁵ L. Labarga,¹⁶ A. Laing,³ P. Lebrun,⁷ I. Liubarsky,³ N. López-March,^{1,3} M. Losada,¹² J. Martín-Albo,² G. Martínez-Lema,¹¹ A. Martínez,³ F. Monrabal,¹ C. M. B. Monteiro,⁹ F. J. Mora,⁸ L. M. Moutinho,⁴ J. Muñoz Vidal,³ M. Musti,³ M. Nebot-Guinot,³ P. Novella,³ B. Palmeiro,³ A. Para,⁷ J. Pérez,³ M. Querol,⁸ J. Repond,¹⁵ J. Renner,³ S. Riordan,¹⁵ L. Ripoll,¹⁷ J. Rodríguez,³ L. Rogers,¹ F. P. Santos,⁵ J. M. F. dos Santos,⁹ A. Simón,³ C. Sofka,^{3,||} M. Sorel,³ T. Stiegler,¹⁸ J. F. Toledo,⁸ J. Torrent,³ Z. Tsamalaidze,¹⁹ J. F. C. A. Veloso,⁴ R. Webb,¹⁸ J. T. White,^{18,*} and N. Yahlali³

(NEXT Collaboration)

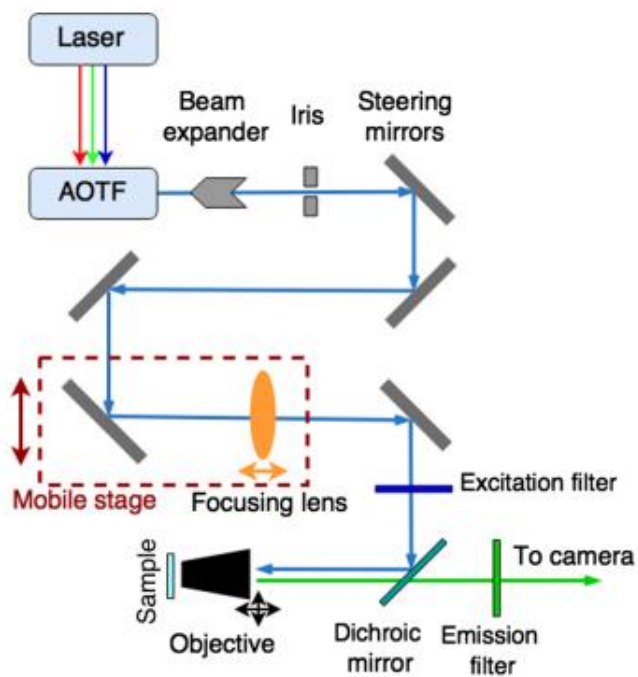


FIG. 2. A schematic view of the TIRF system.

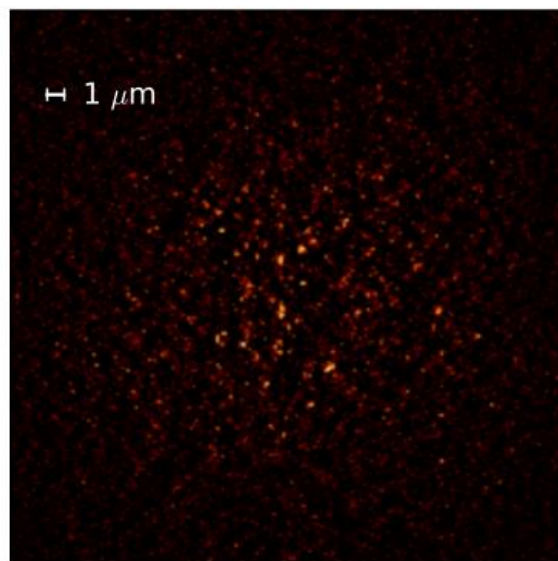


FIG. 3. A sample image from the EM-CCD in one of the barium-spiked samples showing both near-surface (bright) and deeper (dim) fluorescent molecules.

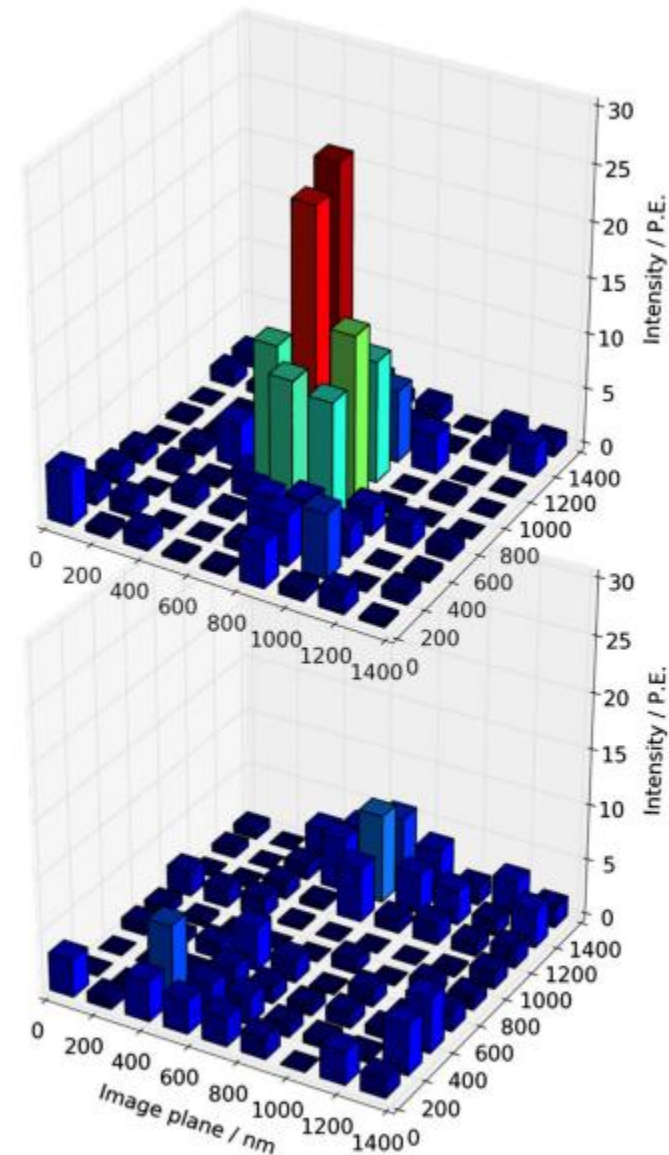
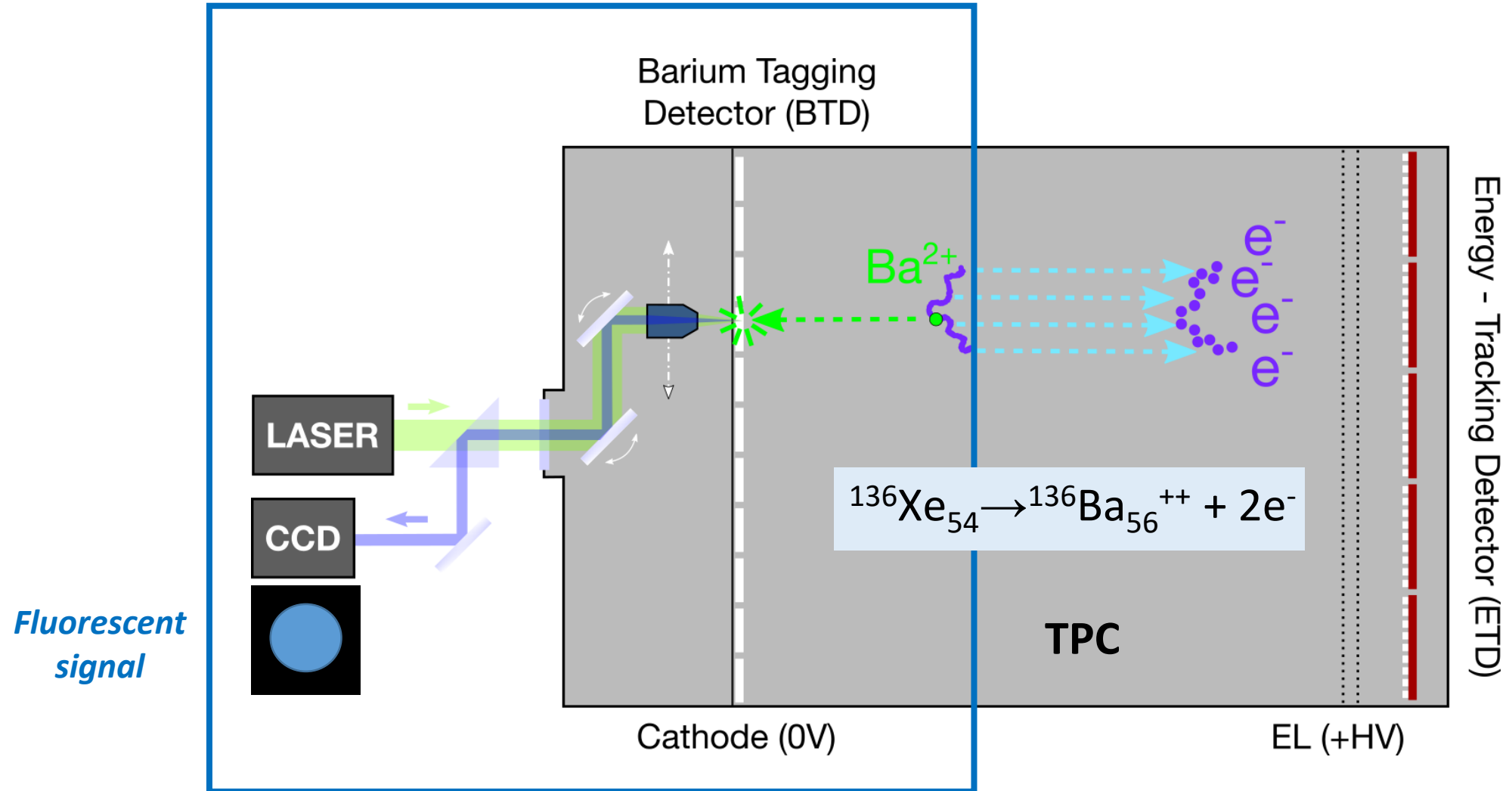


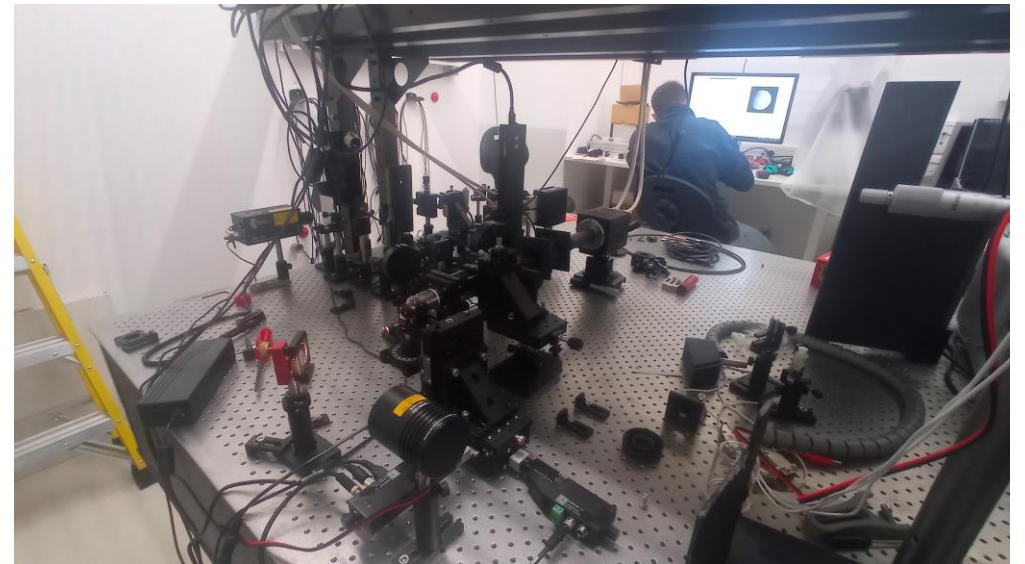
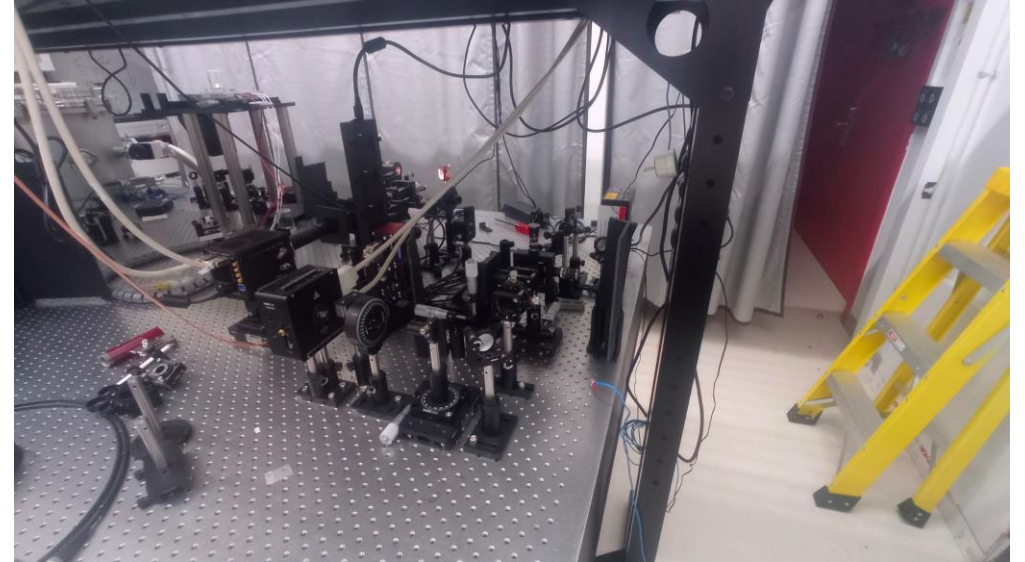
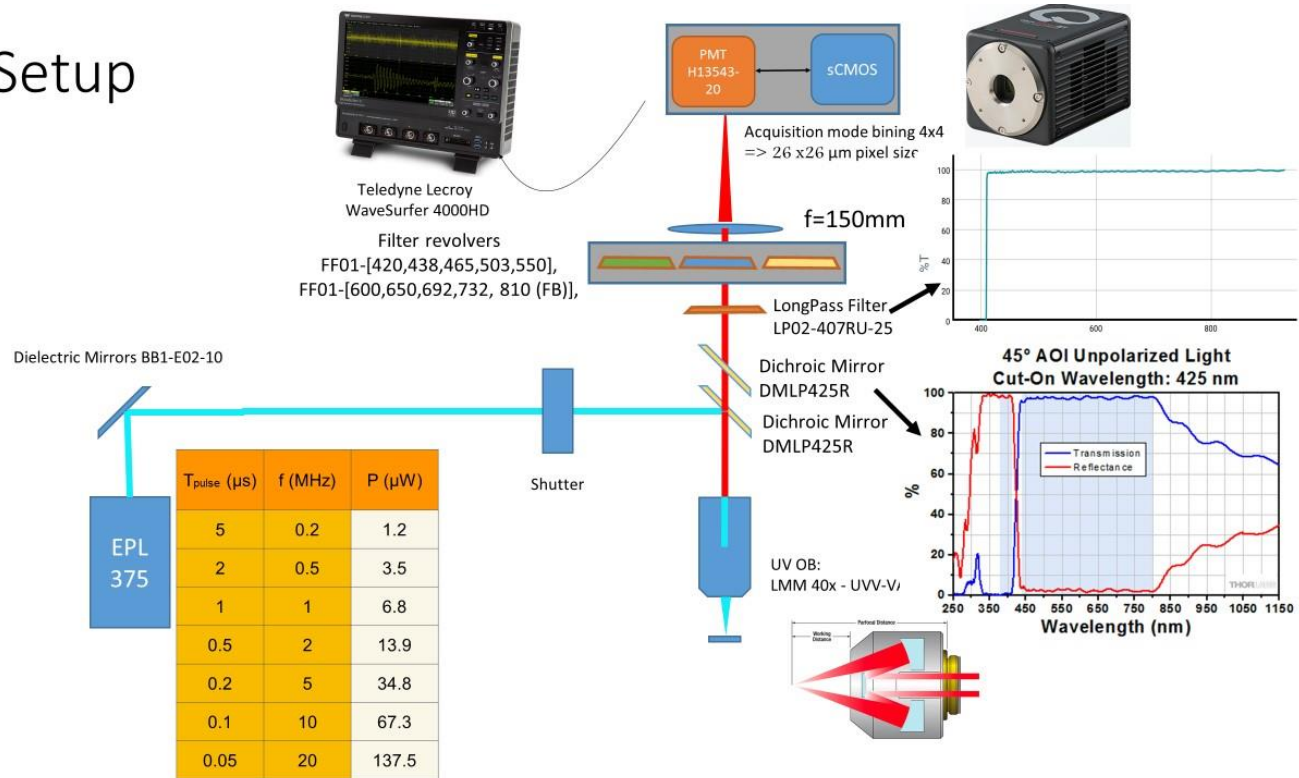
FIG. 1. A single Ba^{++} candidate. A fixed region of the CCD camera is shown with a 0.5 s exposure before (top) and after (bottom) the photobleaching transition.

BOLD/SABAT: The basic scheme

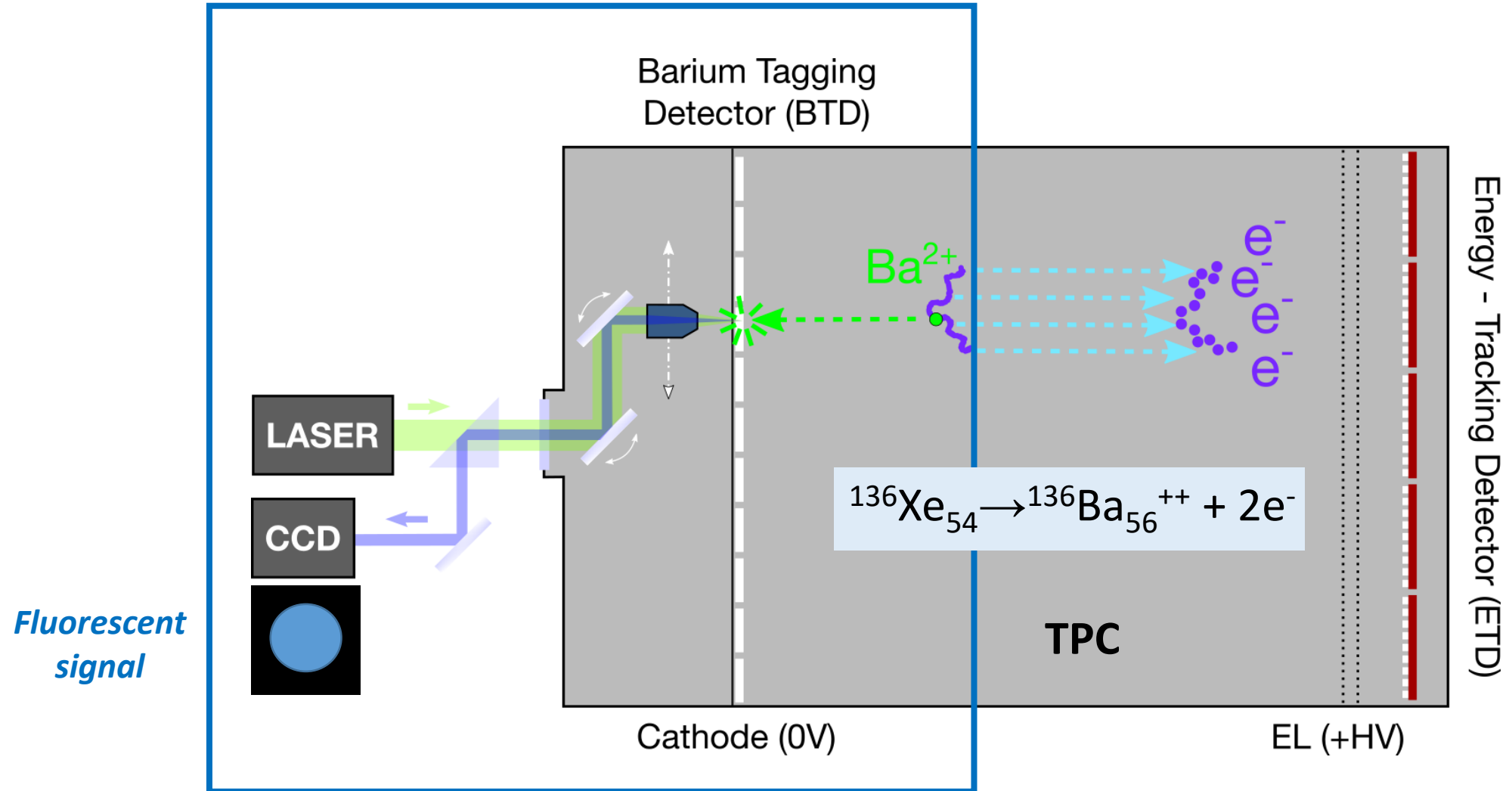


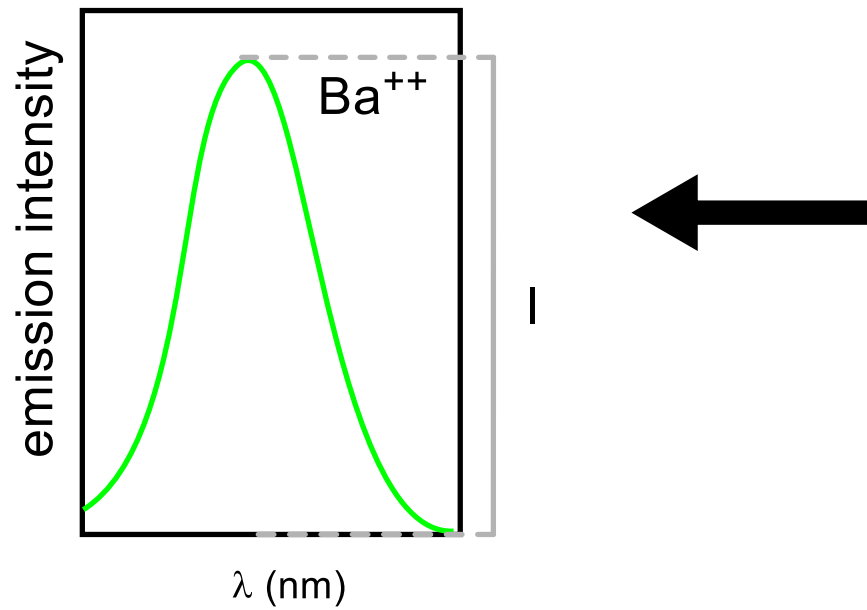
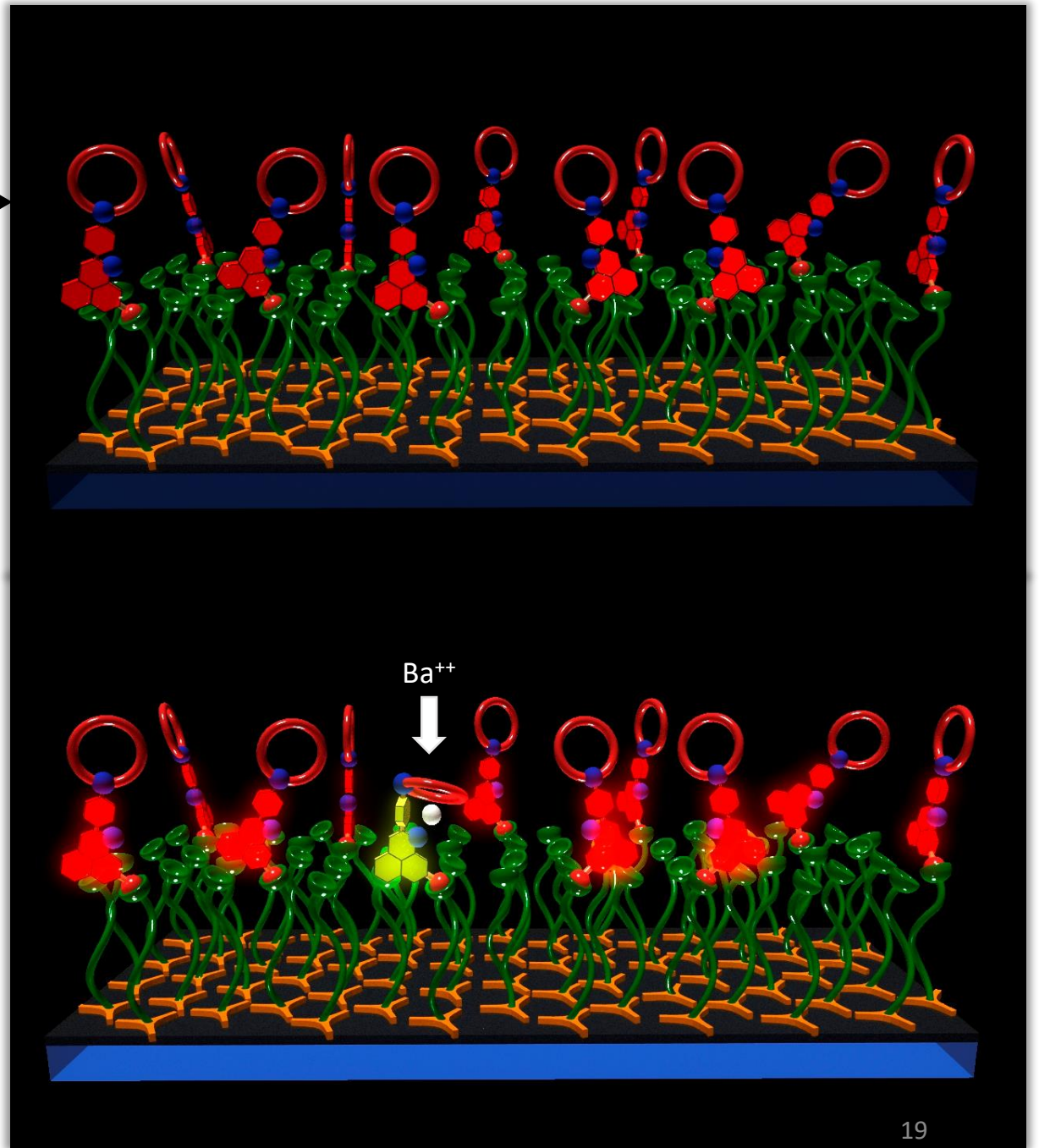
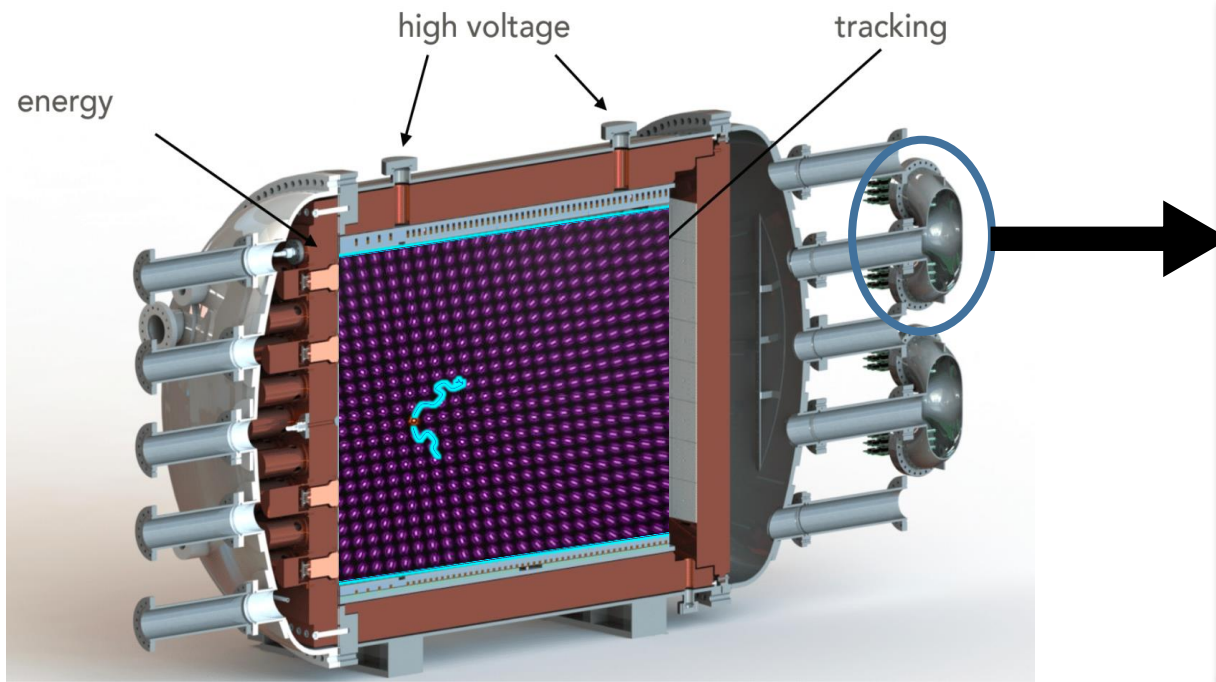
BOLD/SABAT: The basic scheme

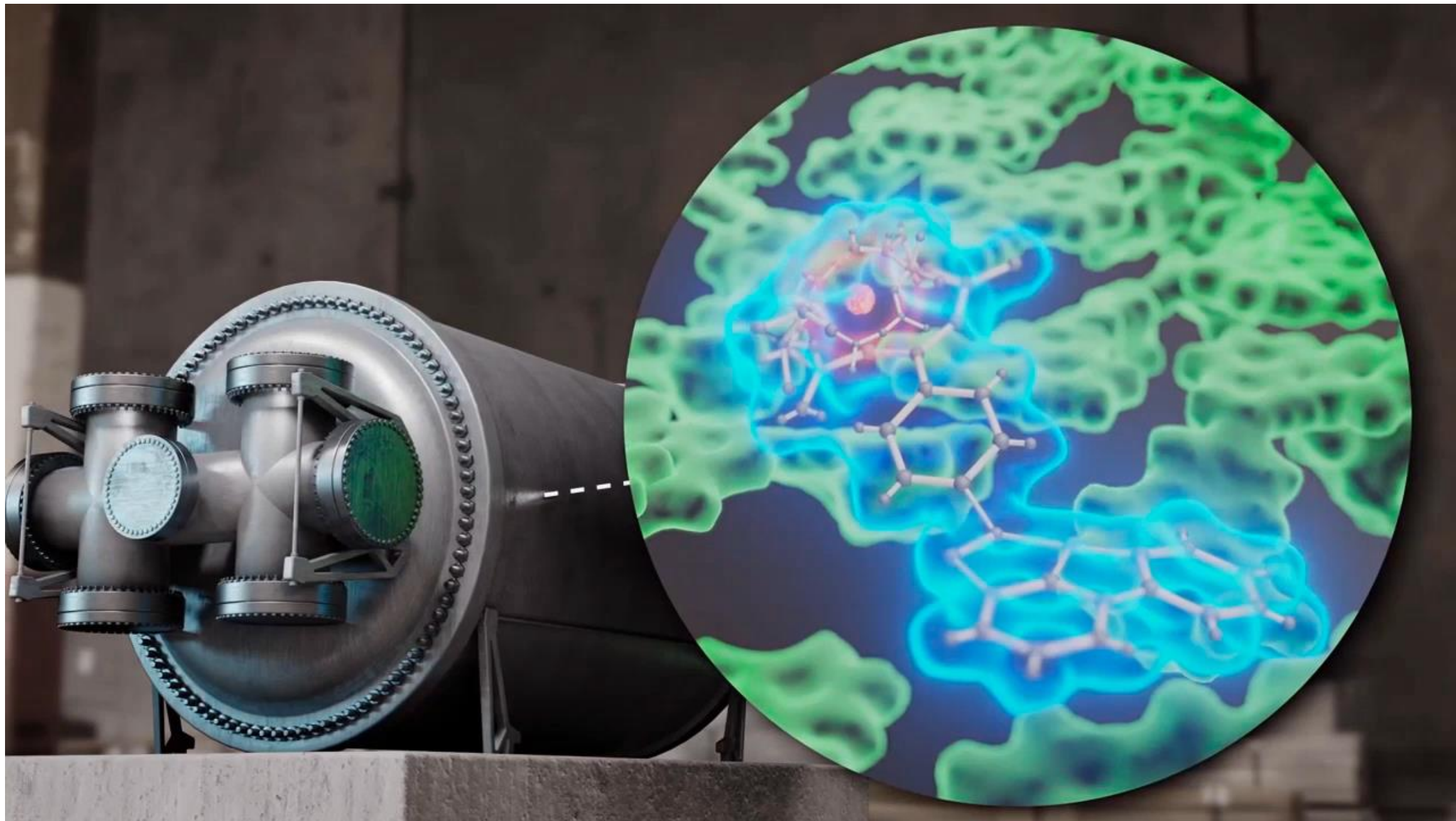
Setup



BOLD/SABAT: The basic scheme





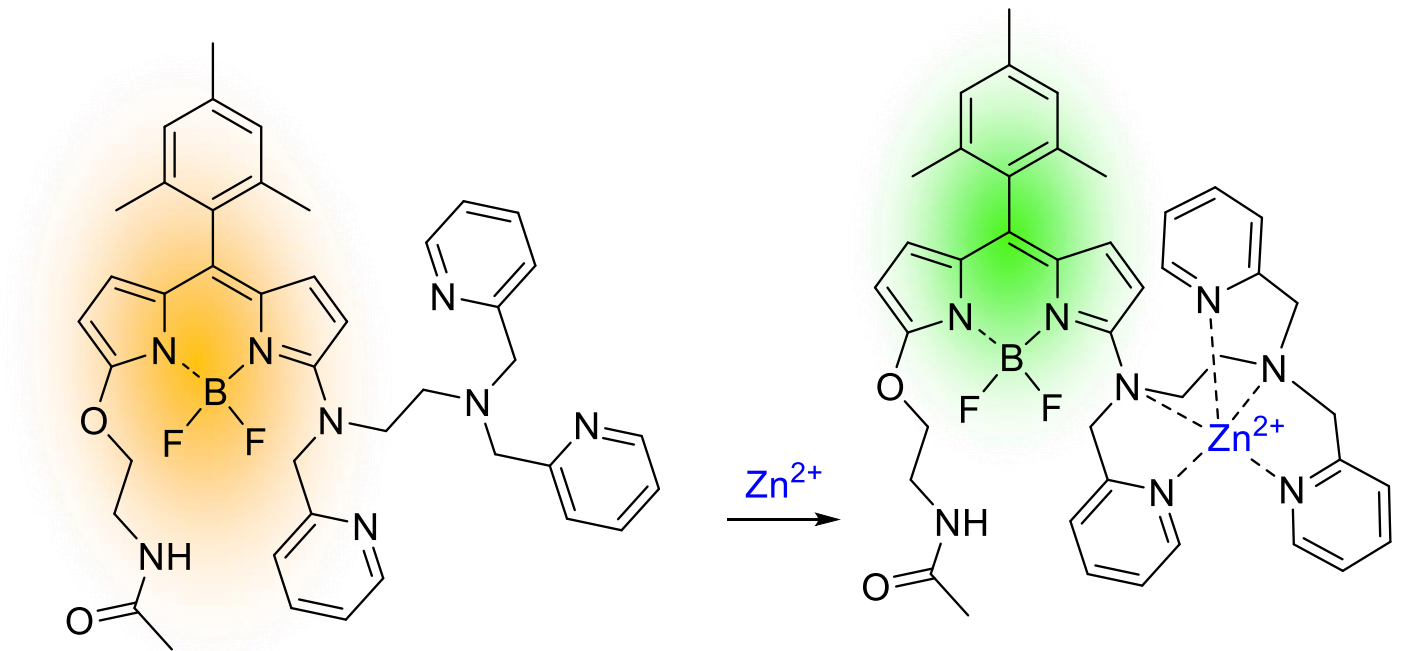


FMI (Fluorescent Monocolor Indicators) and FBI (Fluorescent Bicolor Indicators)

- Dry phase
- Selectivity (Ba^{2+})
- Background free
- 10^{27-28} years

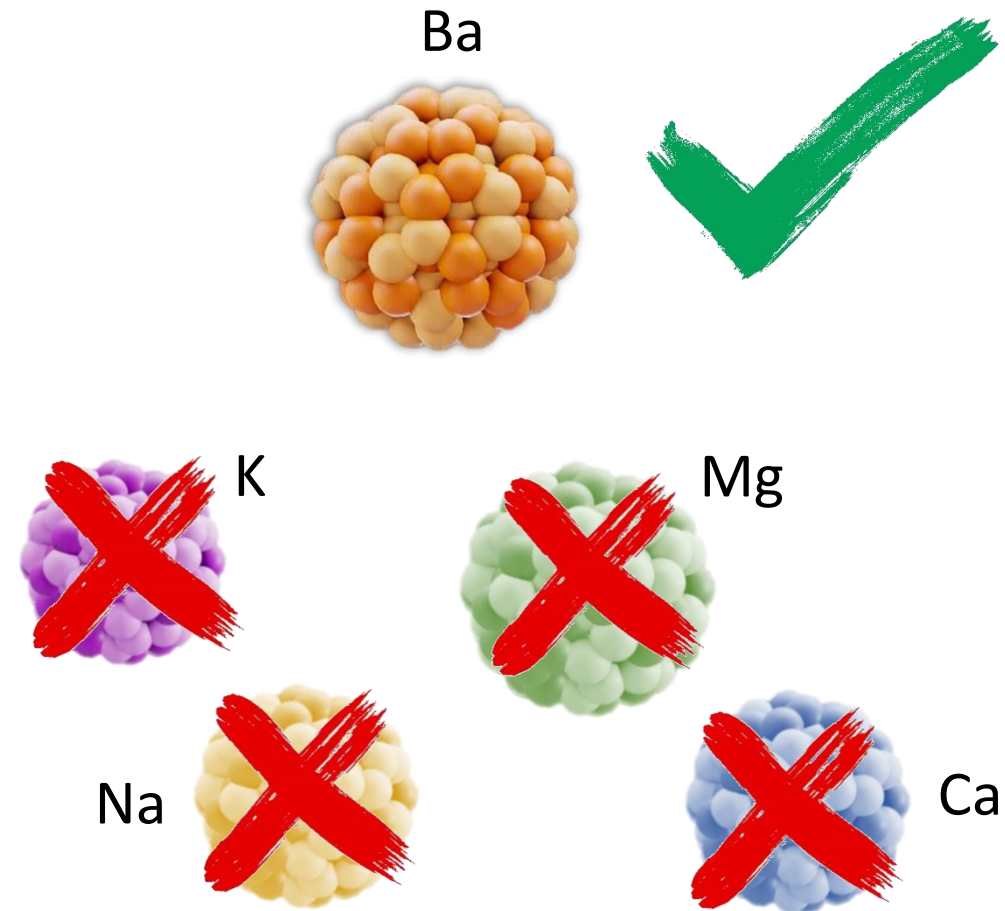
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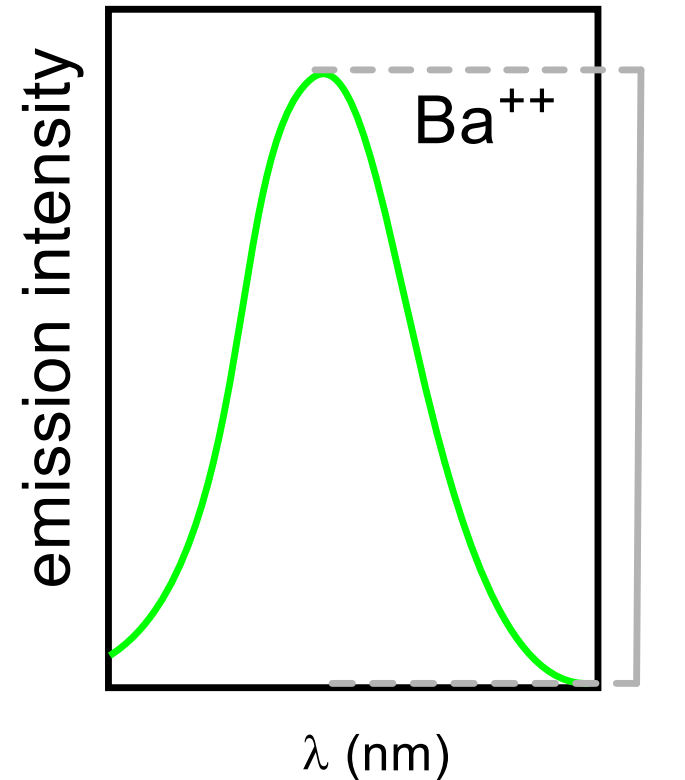
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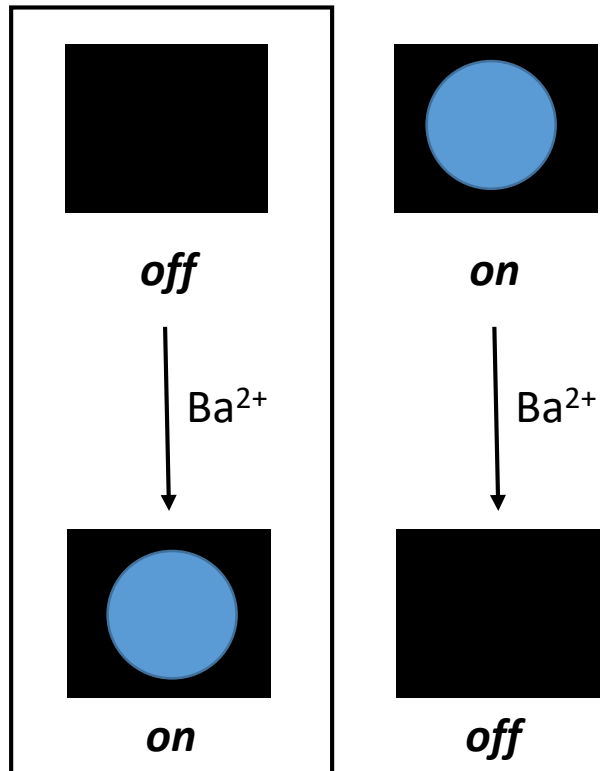


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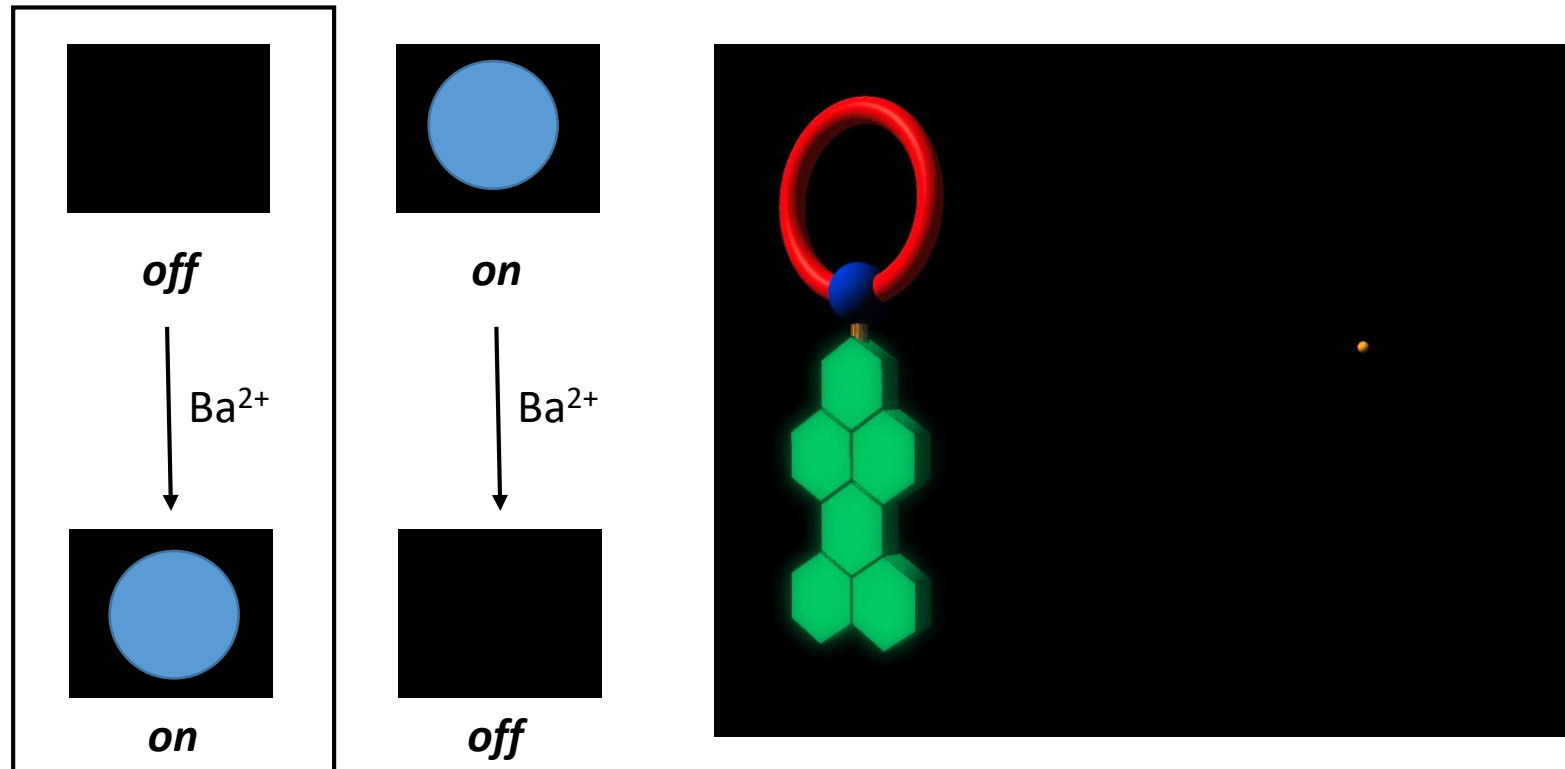


FMI (Fluorescent Monocolor Indicators) and FBI (Fluorescent Bicolor Indicators)



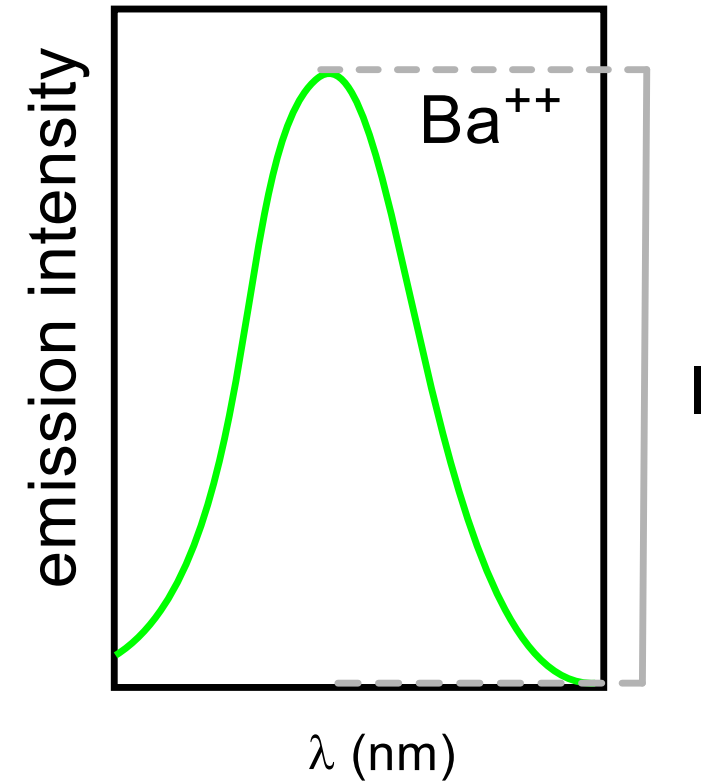
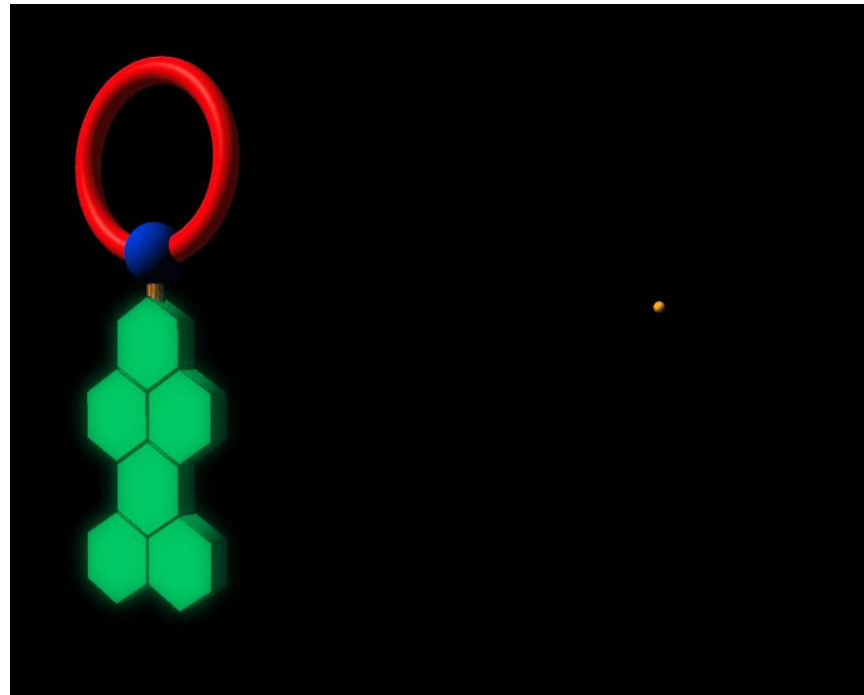
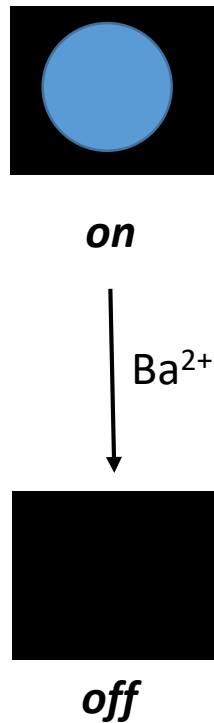
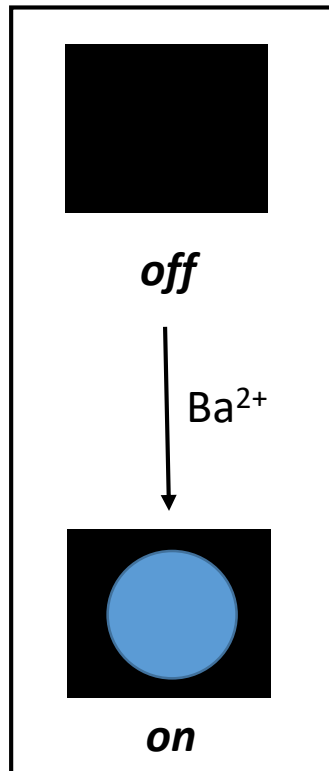
FMIs

FMI (Fluorescent Monocolor Indicators) and FBI (Fluorescent Bicolor Indicators)



FMIs

FMI (Fluorescent Monocolor Indicators) and FBI (Fluorescent Bicolor Indicators)



FMI

FMI (Fluorescent Monocolor Indicators) and FBI (Fluorescent Bicolor Indicators)

Demonstration of Selective Single-Barium Ion Detection with Dry Diazacrown Ether Naphthalimide Turn-on Chemosensors

Pawan Thapa,* Nicholas K. Byrnes,* Alena A. Denisenko, James X. Mao, Austin D. McDonald, Charleston A. Newhouse, Thanh T. Vuong, Katherine Woodruff, Kwangho Nam, David R. Nygren, Benjamin J. P. Jones,* and Frank W. Foss, Jr.*

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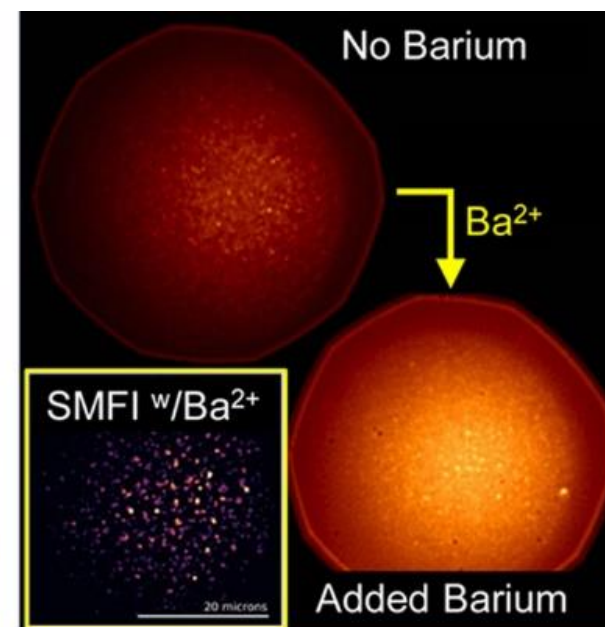
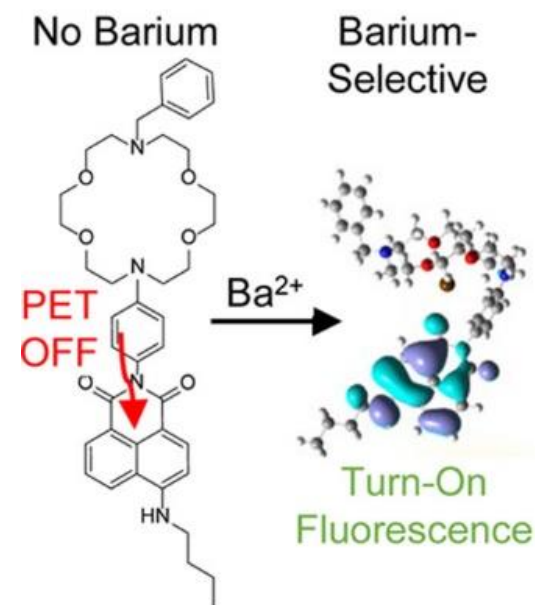
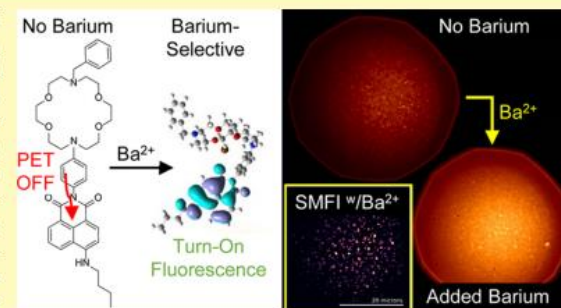
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Supporting Information

ABSTRACT: Single-molecule fluorescence imaging (SMFI) of gas-phase ions has been proposed for “barium tagging,” a burgeoning area of research in particle physics to detect individual barium daughter ions. This has potential to significantly enhance the sensitivity of searches for neutrinoless double-beta decay ($0\nu\beta\beta$) that is obscured by background radiation events. The chemistry required to make such sensitive detection of Ba^{2+} by SMFI in dry Xe gas at solid interfaces has implications for solid-phase detection methods but has not been demonstrated. Here, we synthesized simple, robust, and effective Ba^{2+} -selective chemosensors capable of function within ultrapure high-pressure ^{136}Xe gas. Turn-on fluorescent naphthalimide-(di)azacrown ether chemosensors were Ba^{2+} -selective and achieved SMFI in a polyacrylamide matrix. Fluorescence and NMR experiments supported a photoinduced electron transfer mechanism for turn-on sensing. Ba^{2+} selectivity was achieved with computational calculations correctly predicting the fluorescence responses of sensors to barium, mercury, and potassium ions. With these molecules, dry-phase single- Ba^{2+} ion imaging with turn-on fluorescence was realized using an oil-free microscopy technique for the first time—a significant advance toward single- Ba^{2+} ion detection within large volumes of ^{136}Xe , plausibly enabling a background-independent technique to search for the hypothetical process of $0\nu\beta\beta$.

KEYWORDS: barium tagging, barium sensor, turn-on fluorescence, PET chemosensor, single-molecule imaging



FMI (Fluorescent Monocolor Indicators) and FBI (Fluorescent Bicolor Indicators)

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KEYWORDS: barium tagging, barium sensor, turn-on fluorescence, PET chemosensor, single-molecule imaging

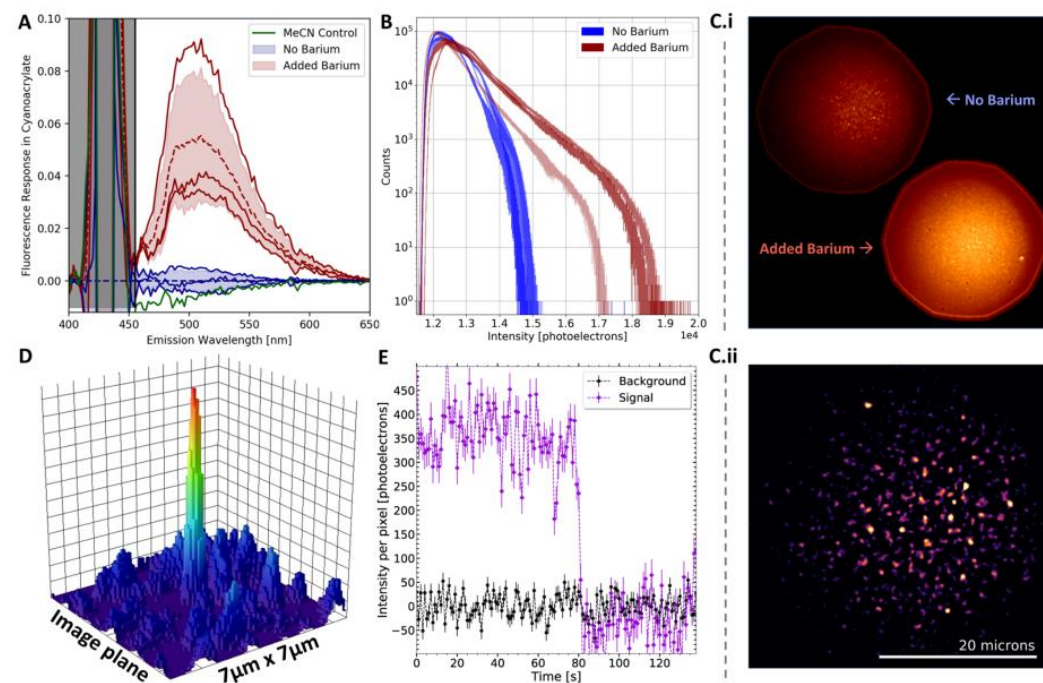
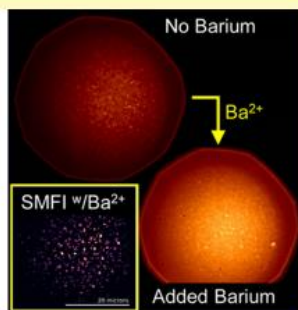
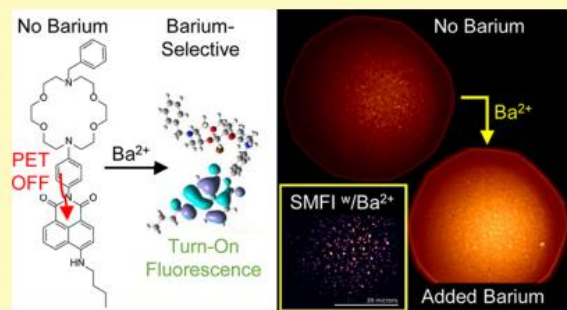
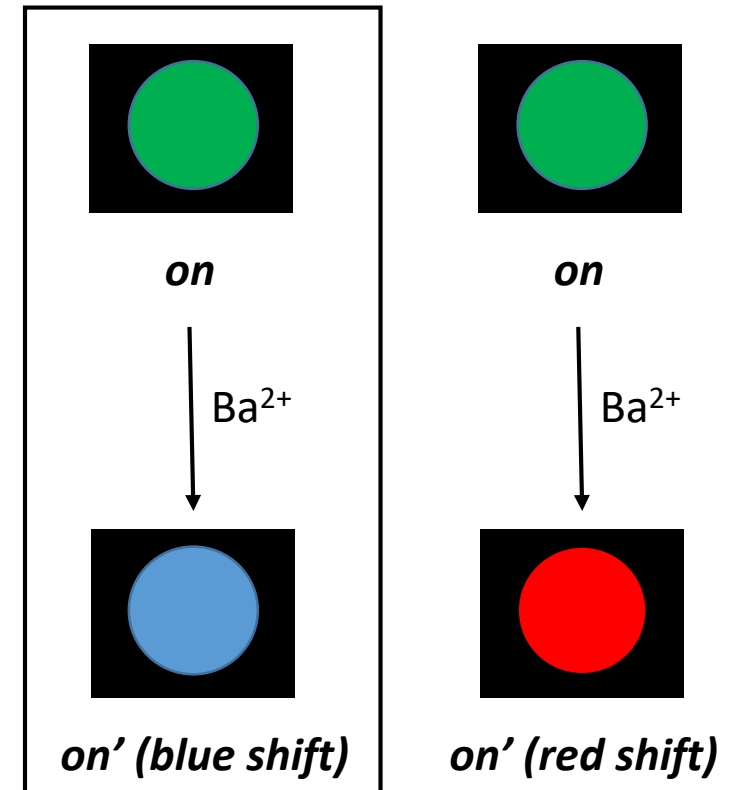


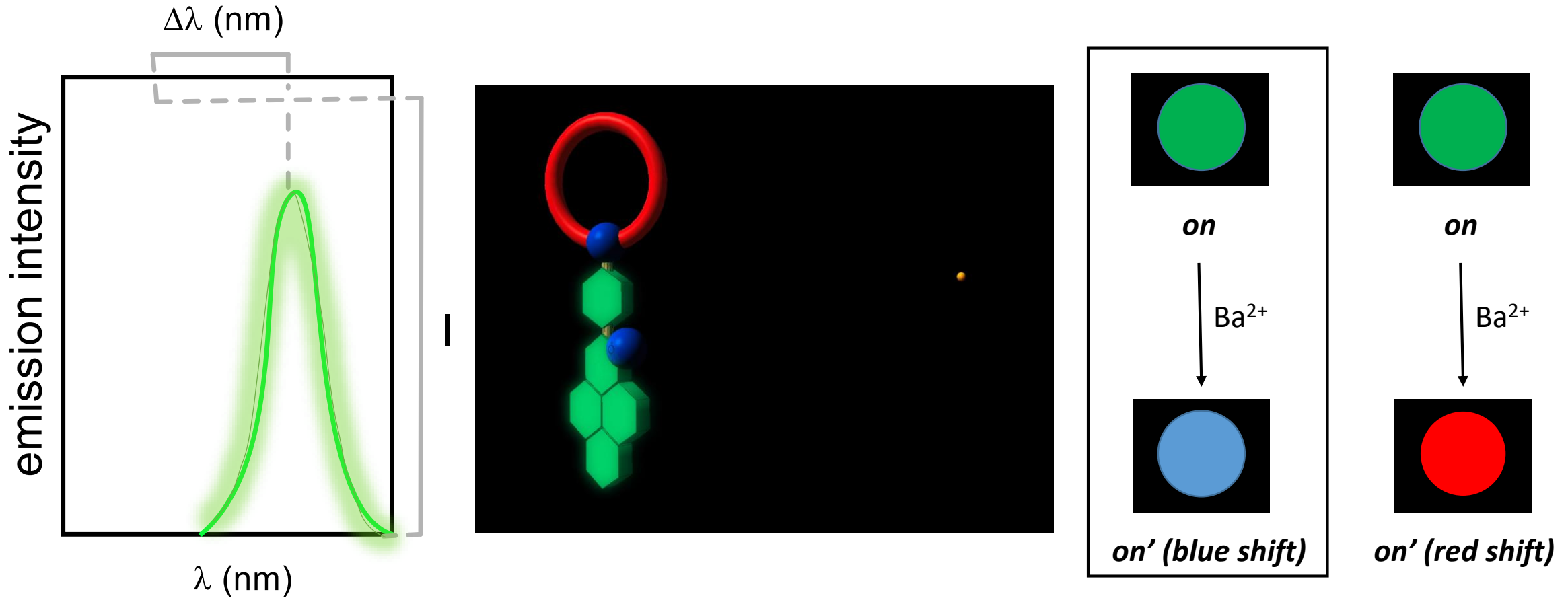
Figure 7. Solid-matrix measurements of barium-sensing fluorophore 1D. (A) Bulk spectrophotometry in a dried cyanoacrylate matrix; (B) raw pixel histogram of three 1D-coated slides imaged at the single-molecule level; (C) single-molecule level microscopy images of the 1D layer with and without Ba^{2+} (i) and with Ba^{2+} after background removal (ii); (D) specific single reconstructed barium-chelated 1D molecule; and (E) barium-chelated 1D molecule fluorescence time trajectory showing a single-step photobleaching characteristic of SMFI detection.

FMI (Fluorescent Monocolor Indicators) and FBI (Fluorescent Bicolor Indicators)



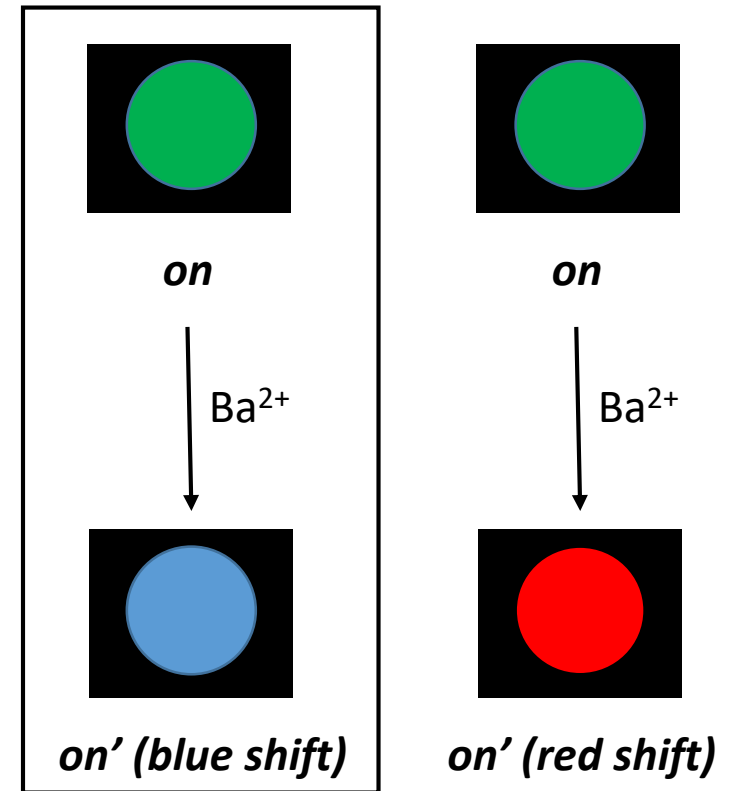
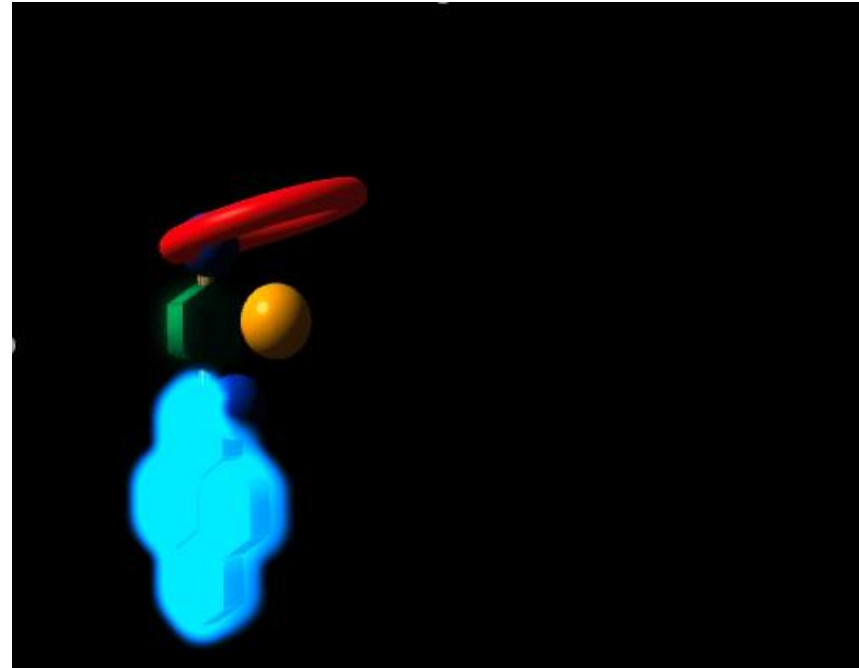
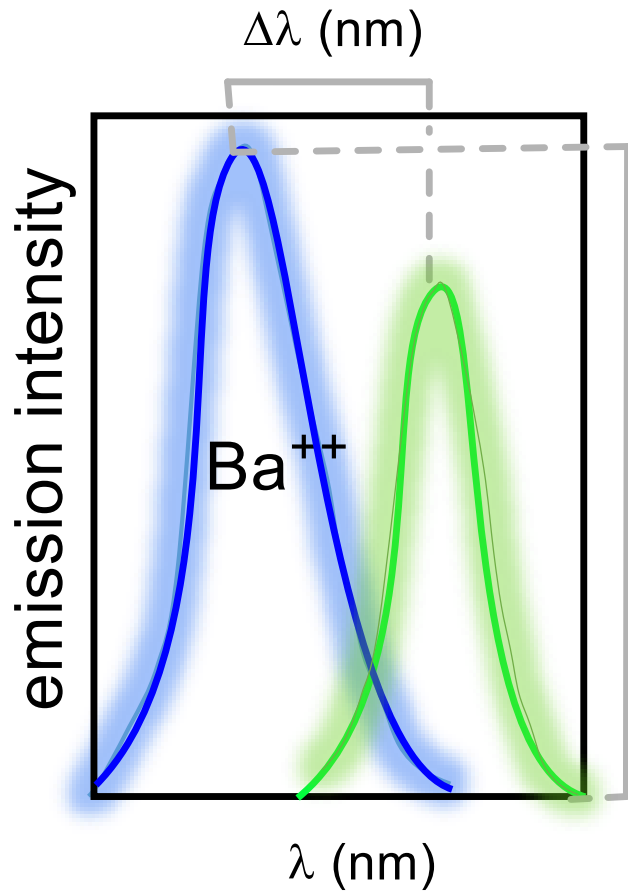
FBIs

FMI (Fluorescent Monocolor Indicators) and FBI (Fluorescent Bicolor Indicators)



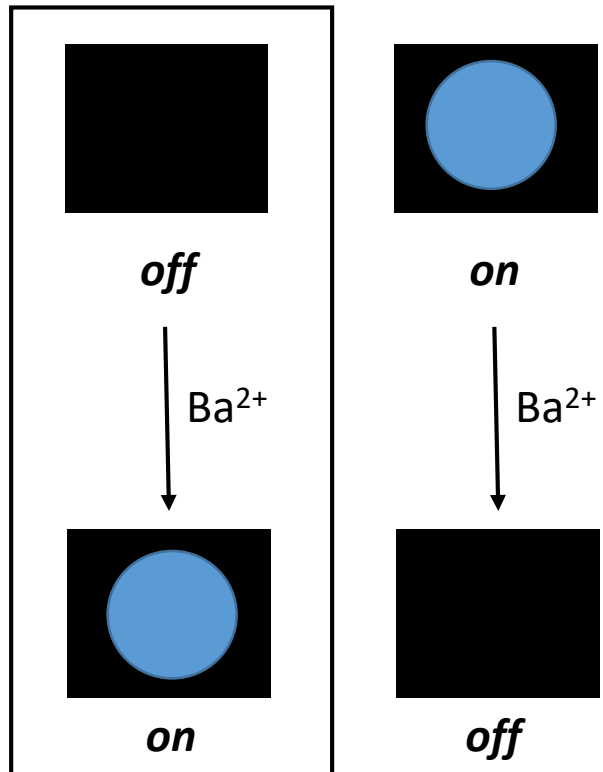
FBIs

FMI (Fluorescent Monocolor Indicators) and FBI (Fluorescent Bicolor Indicators)

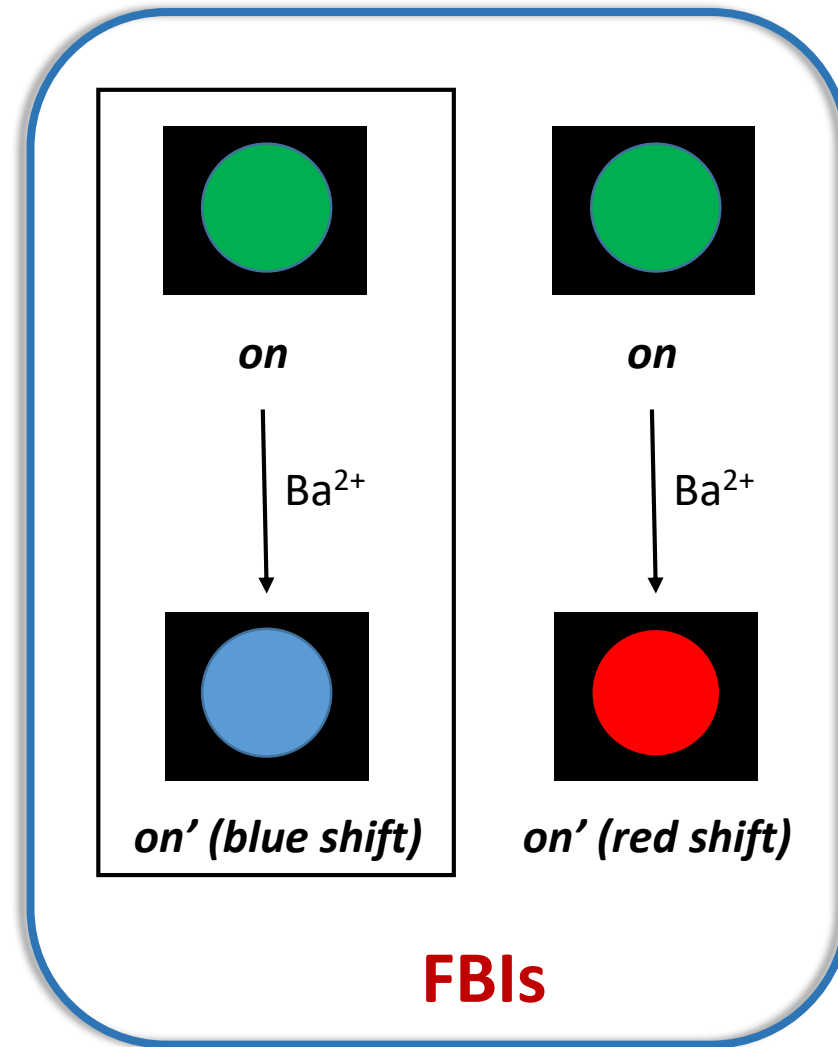


FBIs

FMI (Fluorescent Monocolor Indicators) and FBI (Fluorescent Bicolor Indicators)

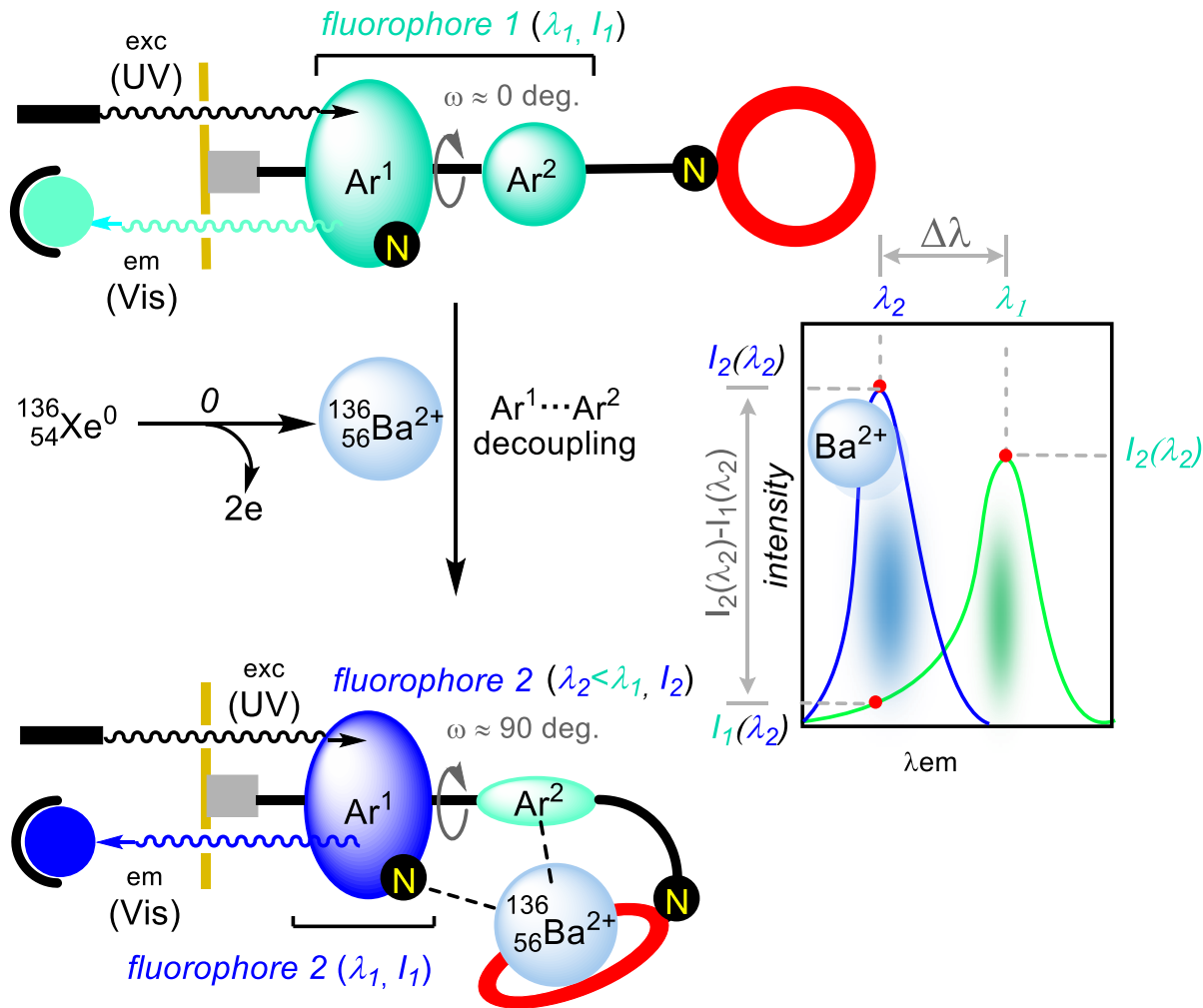


FMIs



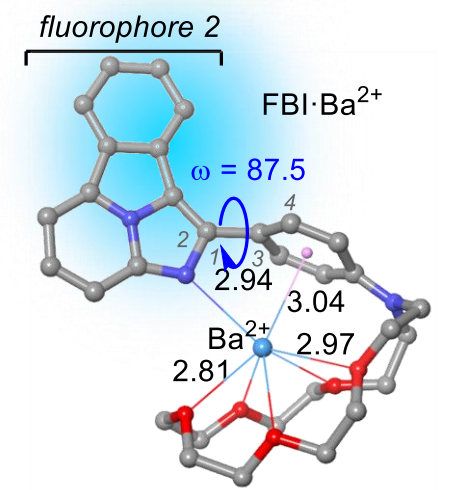
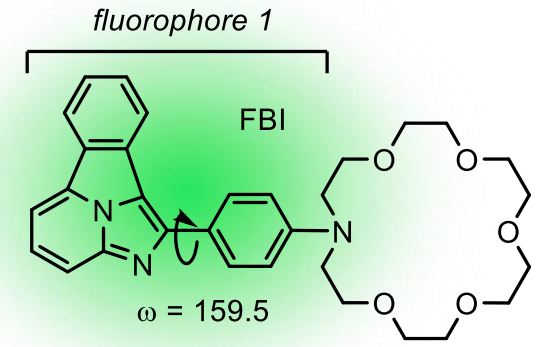
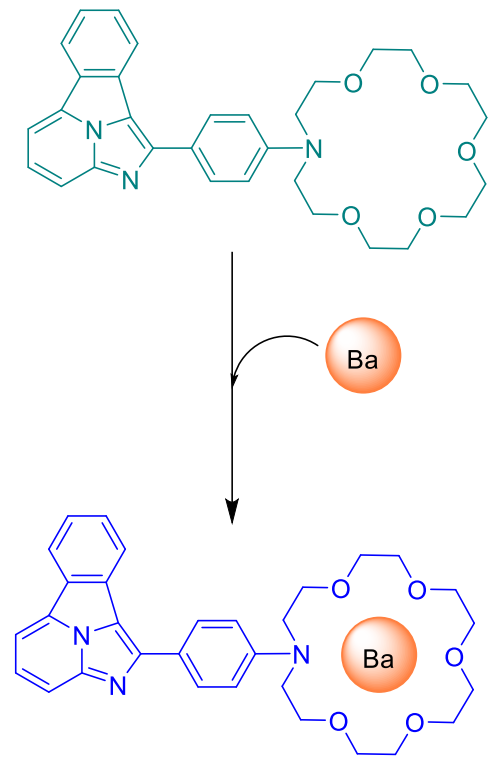
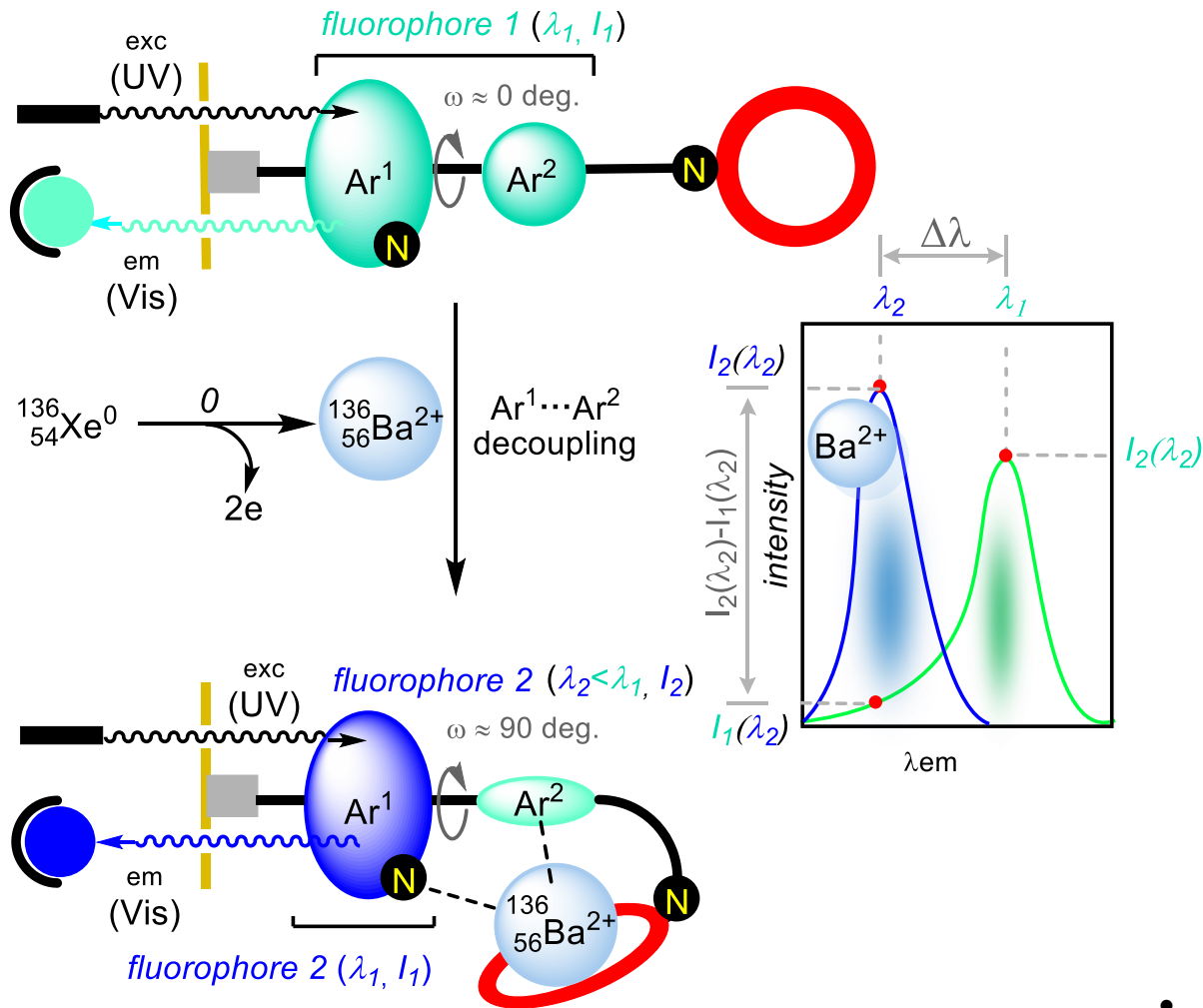
FBIs

1st generation FBIs: Two Fluorophores in One Molecule

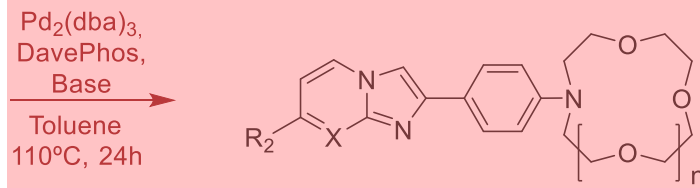
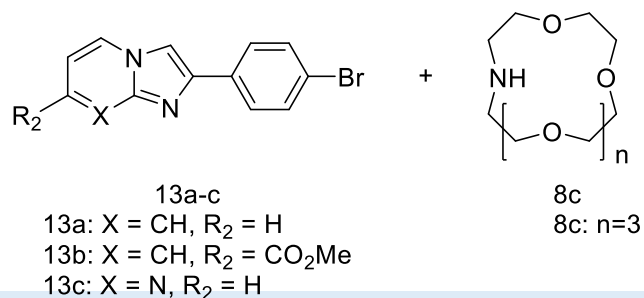


- Two fluorophores (Ph or Hetero-Aromatic-Cyclic system) in one molecule
- Crown Ether (Chelating agent)
- Two responses depending on the coordinator state of the sensor

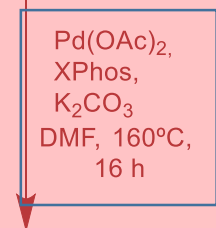
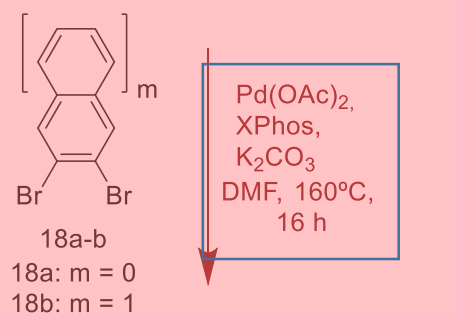
1st generation FBIs: Two Fluorophores in One Molecule



- Green (free)---Blue (Ba⁺⁺)

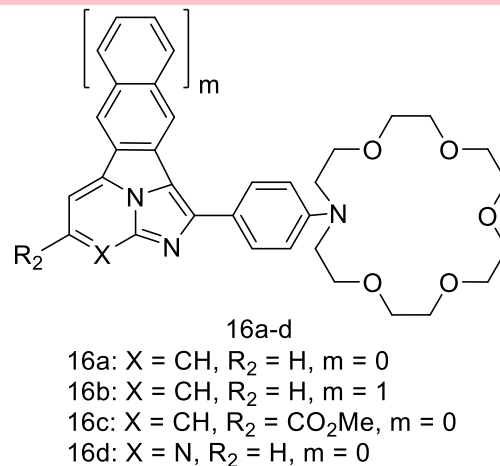
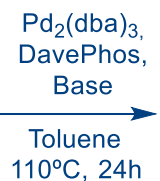
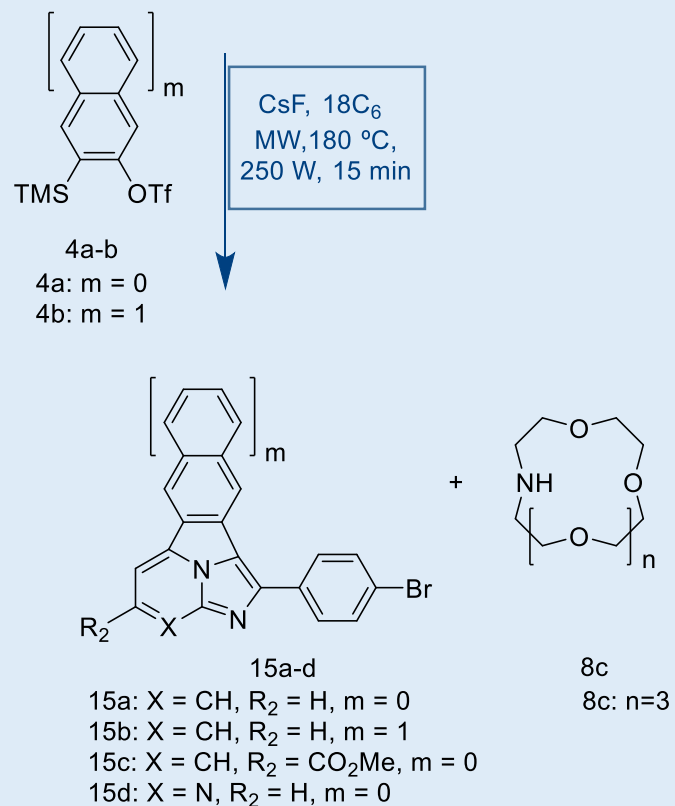


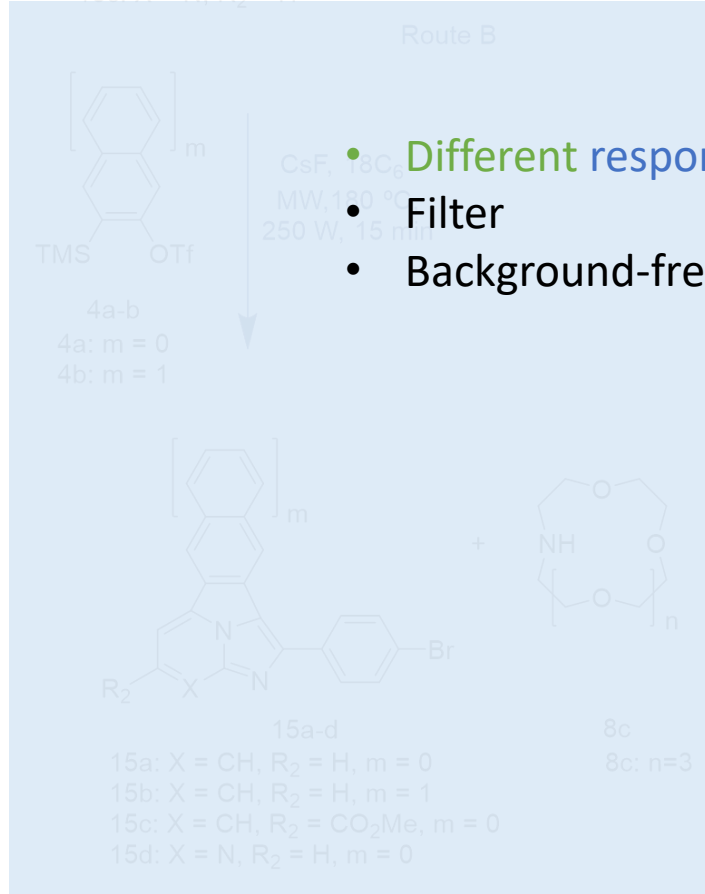
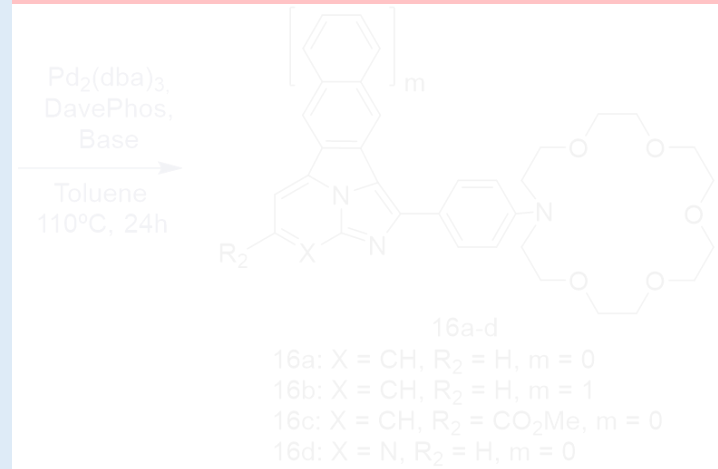
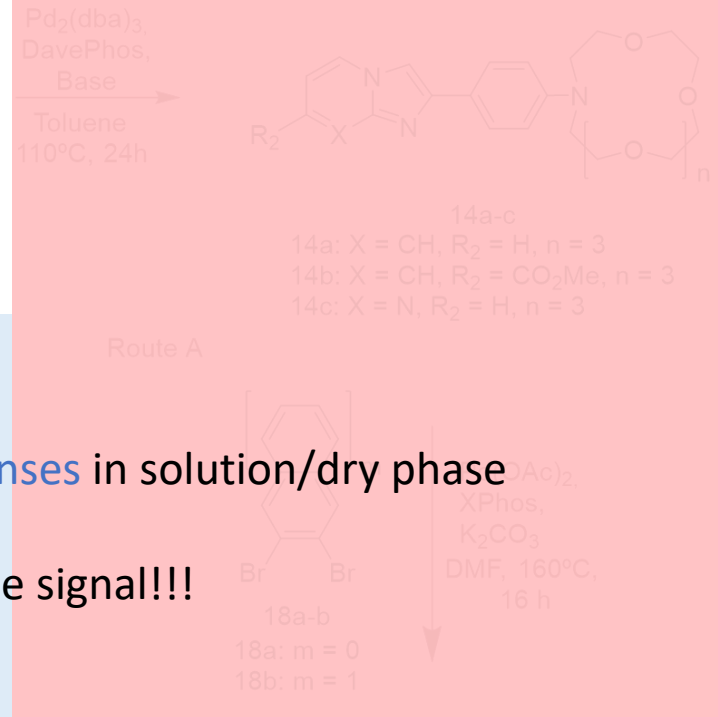
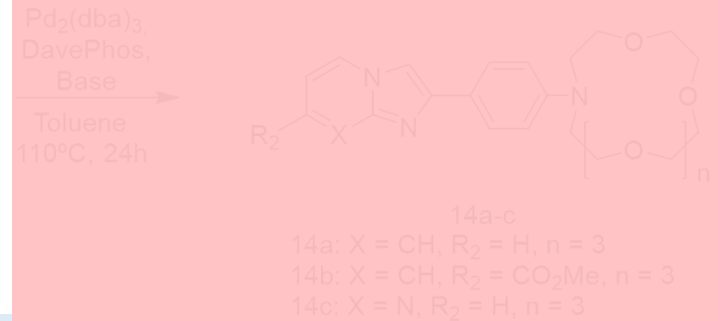
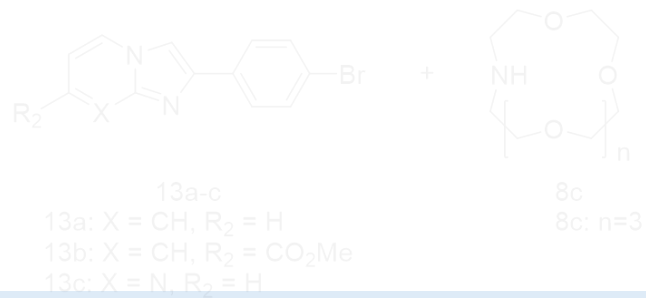
Route A



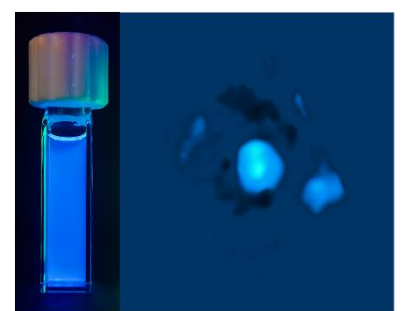
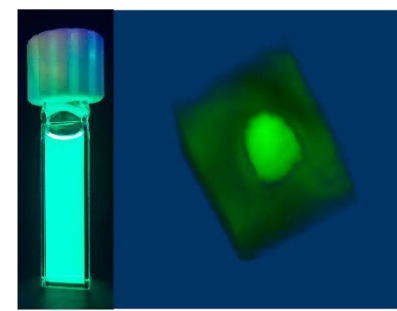
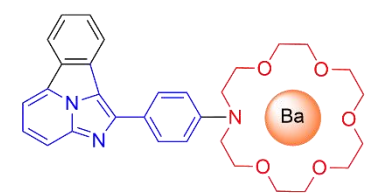
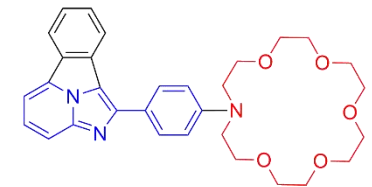
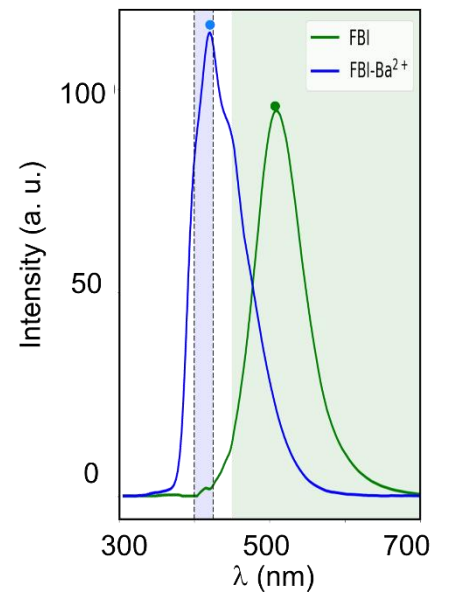
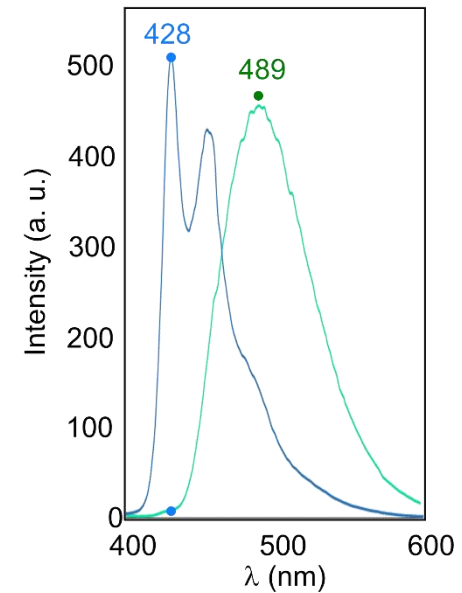
- C-C or C-N coupling reactions
- High order 8+2 cycloadditions reactions
- Library of sensors or candidates

Route B





- Different responses in solution/dry phase
- Filter
- Background-free signal!!!



Article

Fluorescent bicolour sensor for low-background neutrinoless double β decay experiments

<https://doi.org/10.1038/s41586-020-2431-5>

Received: 15 September 2019

Accepted: 3 April 2020

Published online: 22 June 2020

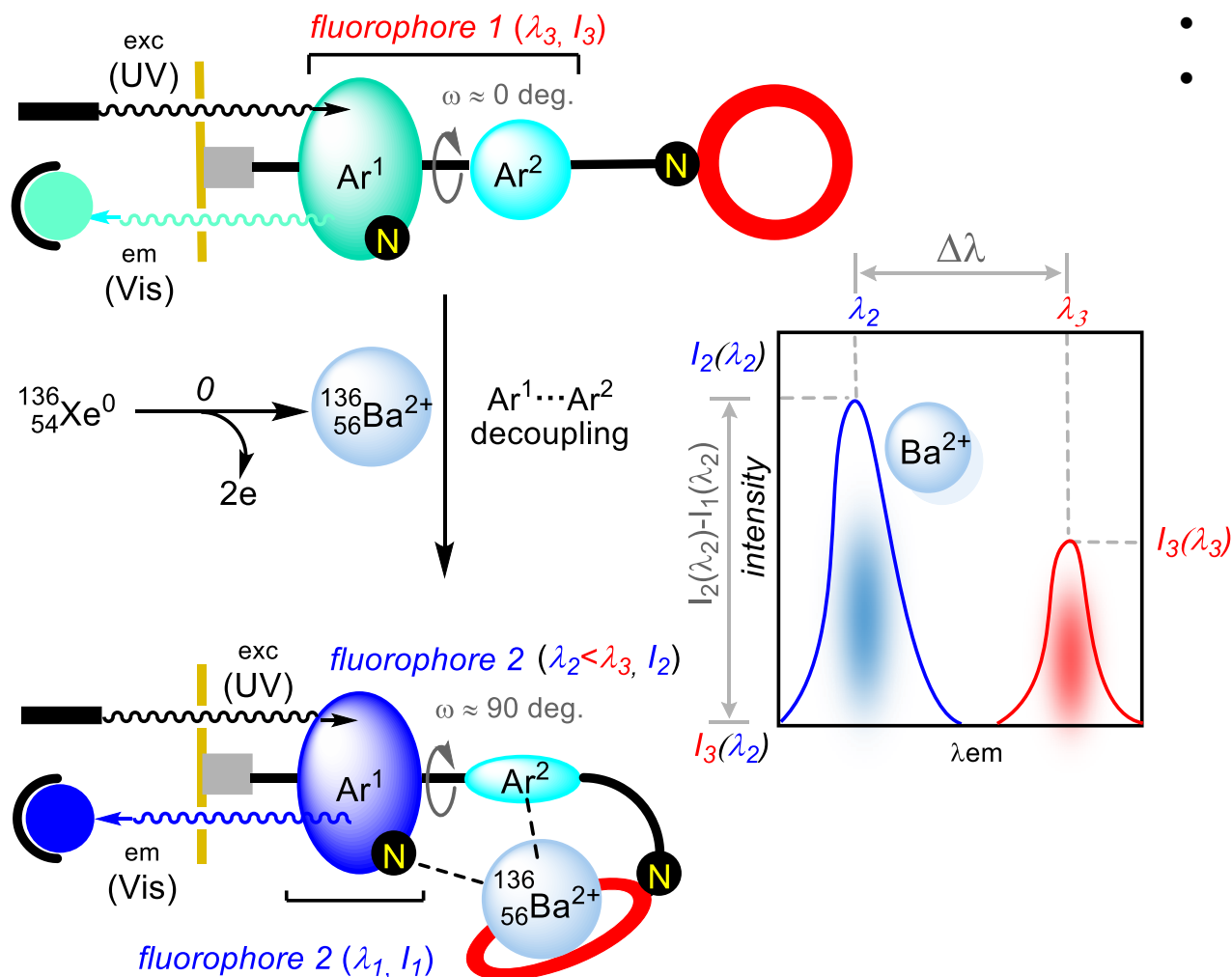
Iván Rivilla¹, Borja Aparicio², Juan M. Bueno³, David Casanova^{1,4}, Claire Tonnelé¹, Zoraida Freixa^{4,5}, Pablo Herrero¹, Celia Rogero^{1,6}, José I. Miranda⁷, Rosa M. Martínez-Ojeda³, Francesc Monrabal^{1,4}, Beñat Olave⁸, Thomas Schäfer^{4,8}, Pablo Artal³, David Nygren⁹, Fernando P. Cossío^{1,2}✉ & Juan J. Gómez-Cadenas^{1,4}✉

https://www.youtube.com/watch?v=W4_Qjd3h2PU

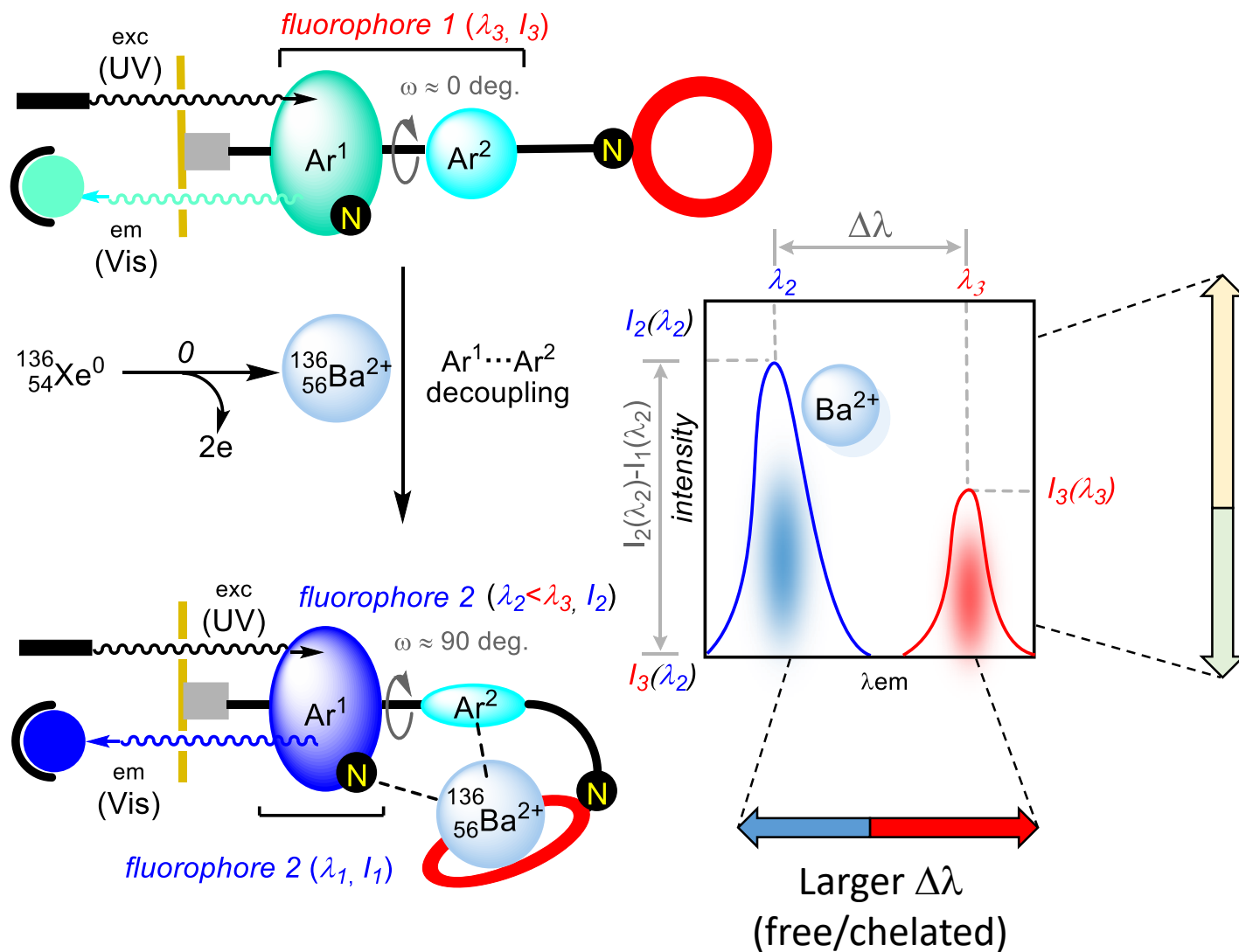


Optimizing the photophysics of FBIs: Design Criteria for Next Generations

- ...a lot of room for further improvement!!!
- Strict rules...NEXT/BOLD experiments
- The pipeline pharma strategy



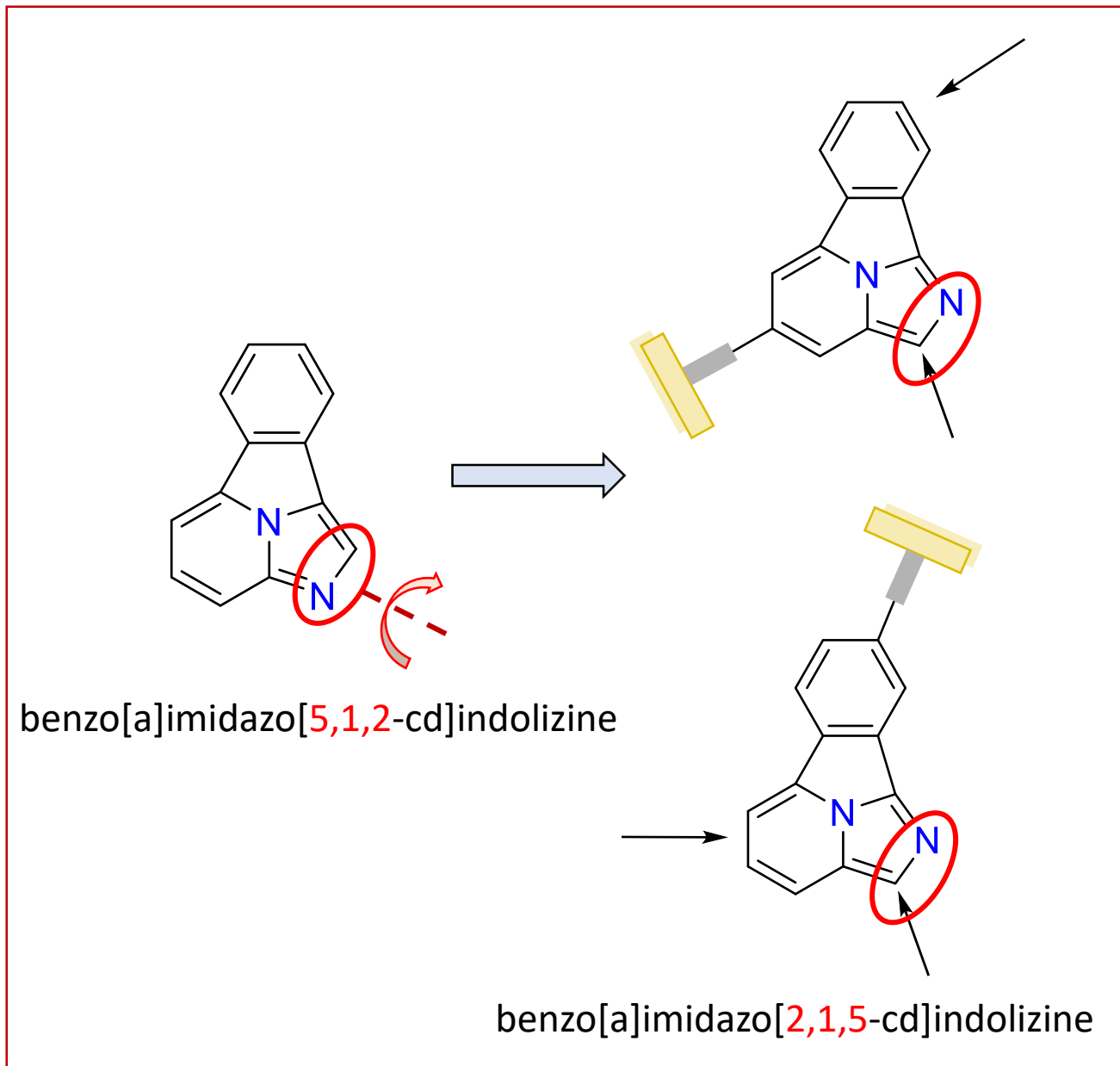
Optimizing the photophysics of FBIs: Design Criteria for Next Generations



To increase (Neutrinoless $\beta\beta$ extremely rare event)

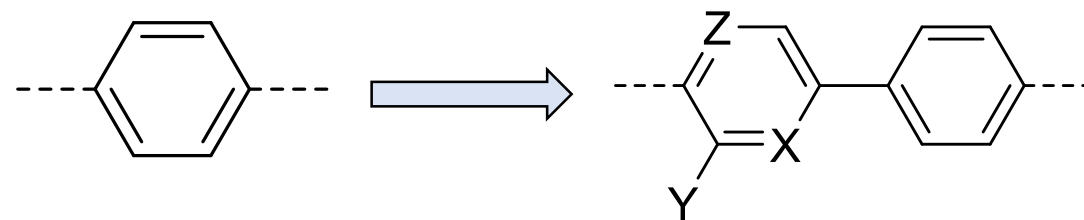
- Higher brightness
- Larger discrimination factor
- Bigger binding constant

Next generation FBIs: Novel Ar¹ and Ar² Units



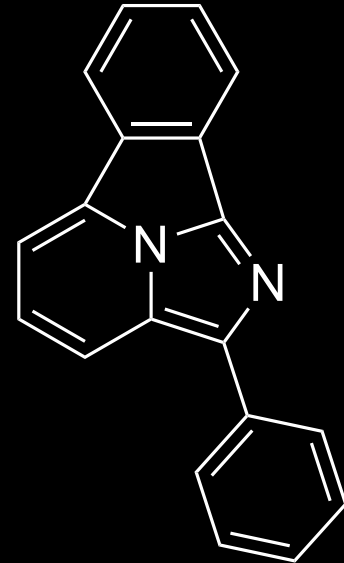
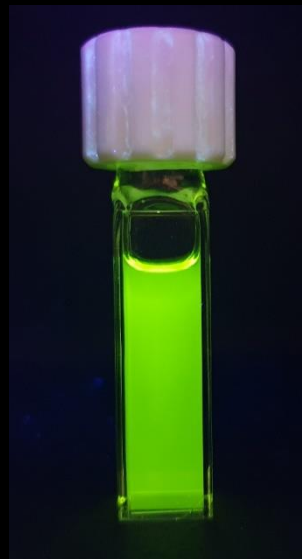
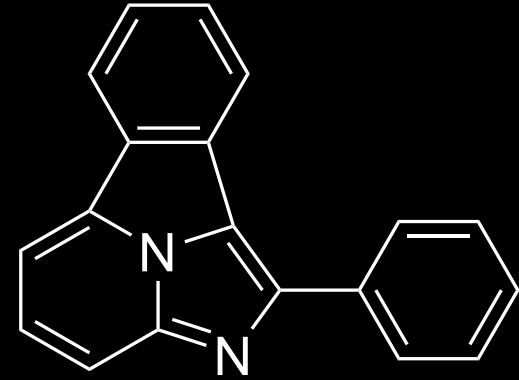
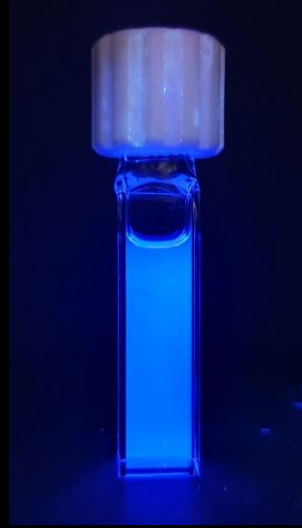
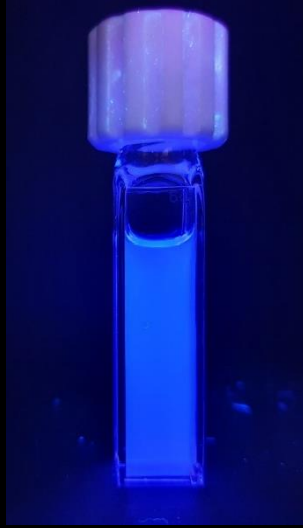
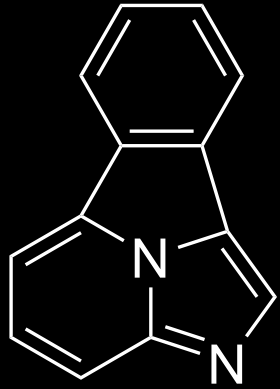
Different candidates (Design)

- Rotation ... changing the nitrogen by carbon,
- 5,1,2 to 2,1,5
- Configurational space
- Decorate options
- Anchor to surfaces

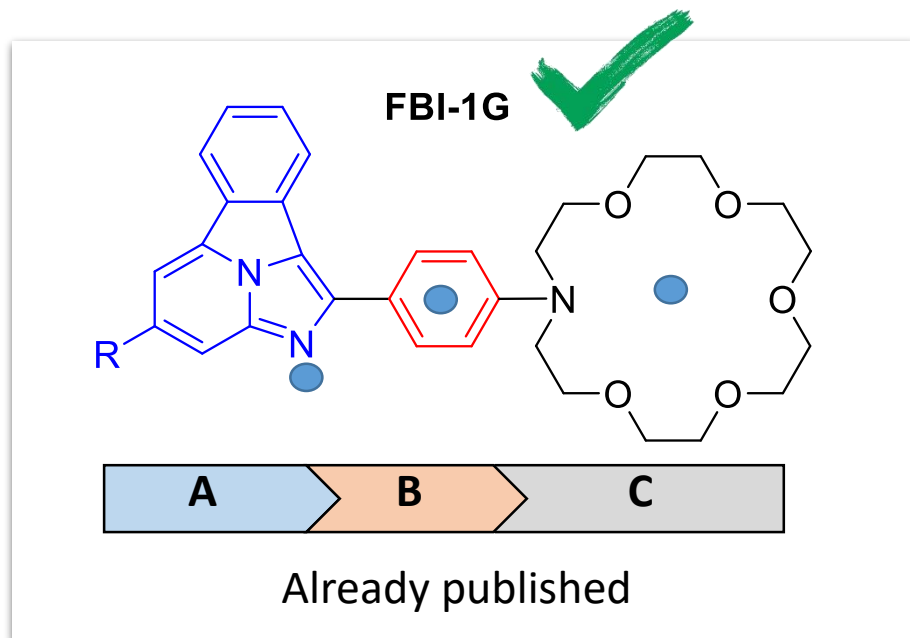


- a: X=Z=CH; Y=H
 b: X=N; Y=CH₃; Z=CH
 c: X=Z=N; Y=H

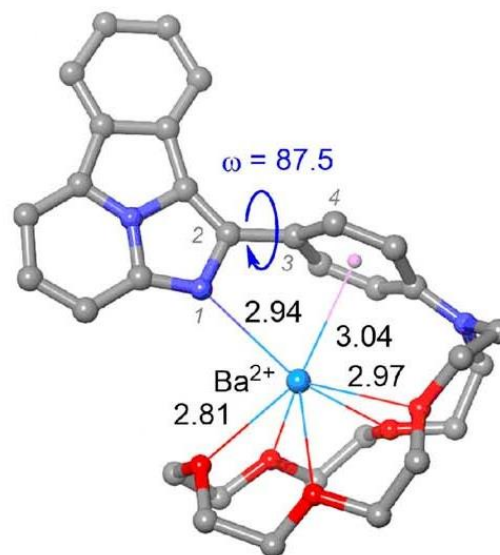
$\lambda_{\text{exc}} = 365 \text{ nm}$
[] = $5 \cdot 10^{-5} \text{ M}$
ACN



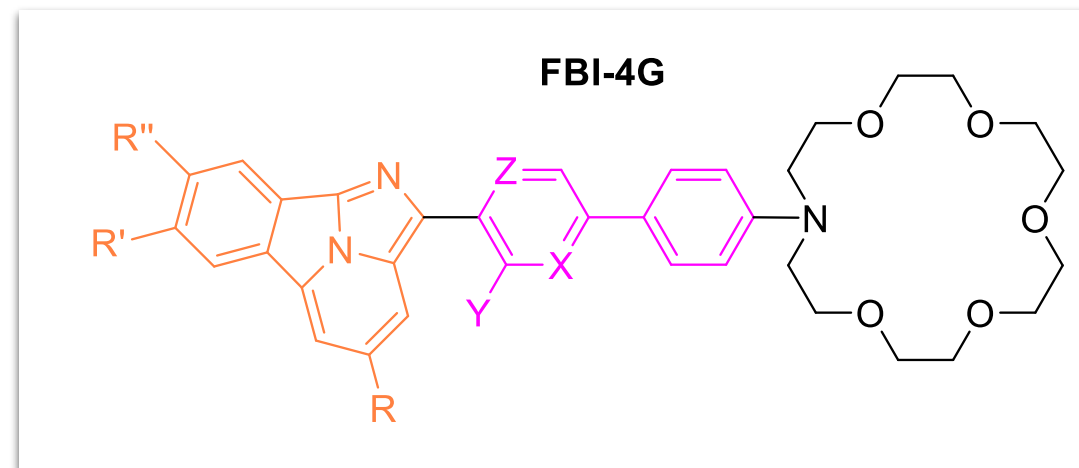
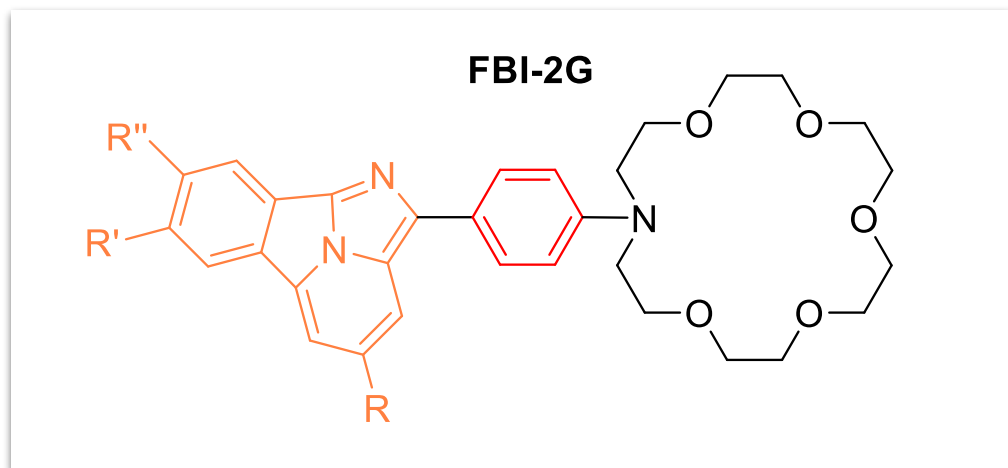
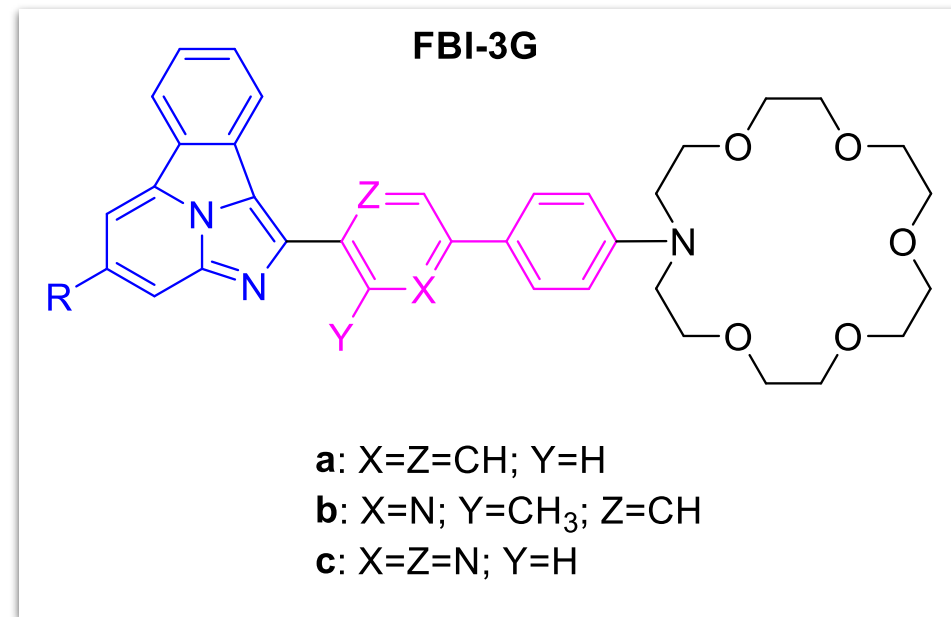
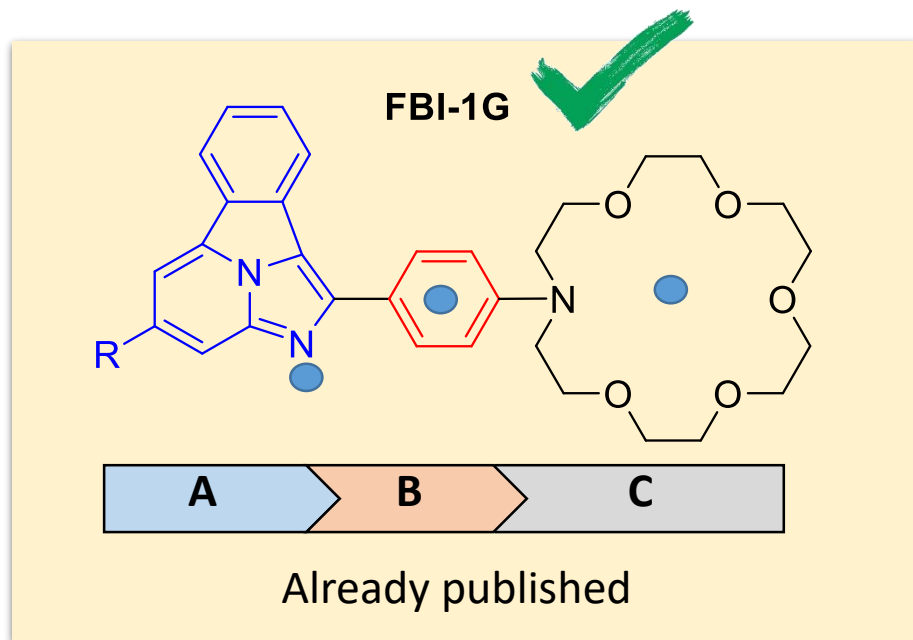
FBI Series: Four Generations



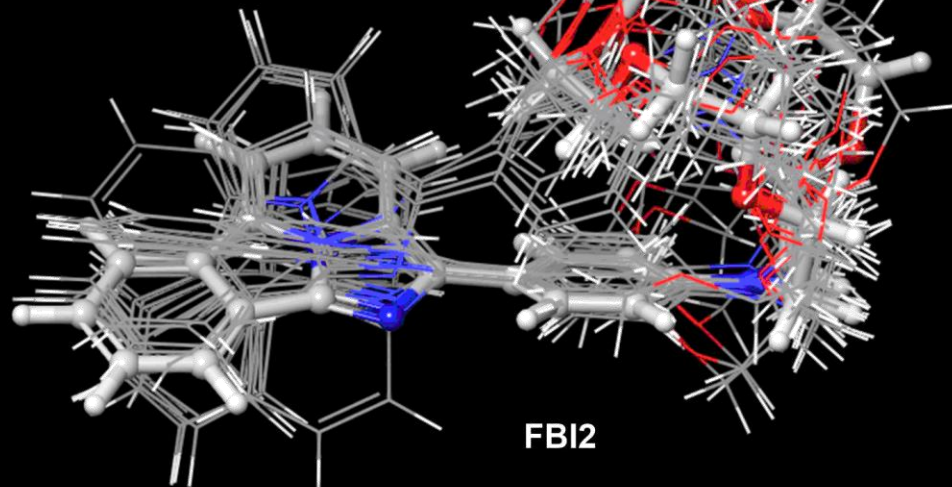
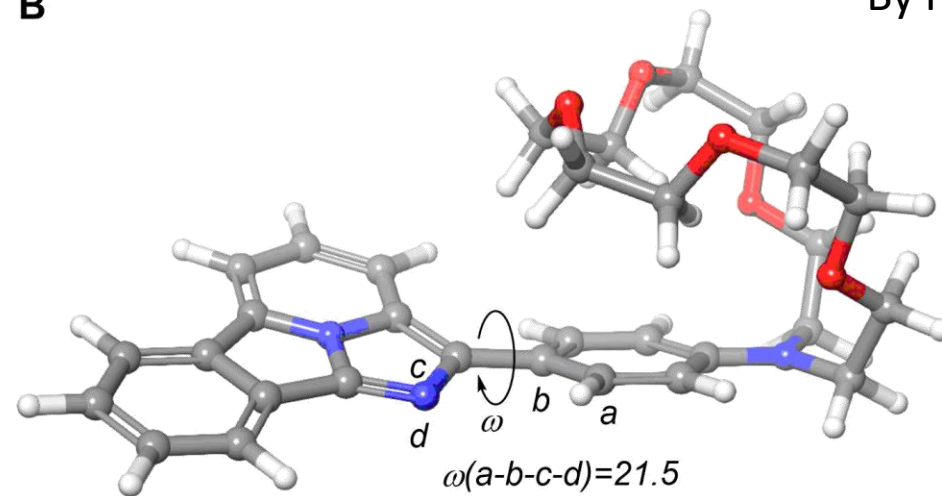
- 3 components (**A**, **B** and **C**)
- **A** Heterocycle; **B** Phenyl group and **C** Crown Ether
- Interaction with Ba²⁺
- Center of Crown Ether
- Pi-cation-interaction-aromatic ring
- N-from heterocycle



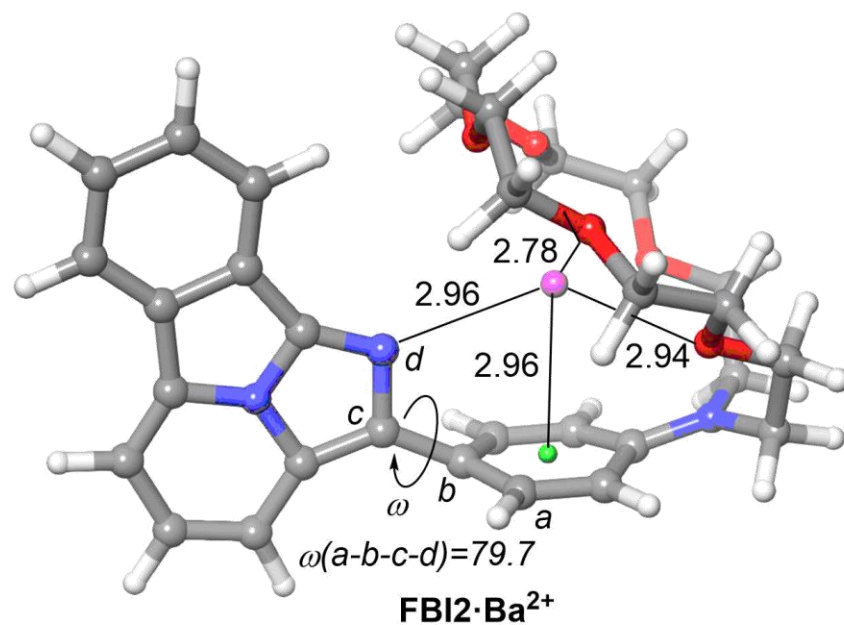
FBI Series: Four Generations



A Force field: OPLS3e
 $\Delta E(\text{MM})=4.0 \text{ kJ/mol}$
 #Structures=18

**B****FBI2**

- Strong conformational flexibility
- Phenyl group coplanar with heterocycle (G1)
- Rigid conformation than G1 (Ba++)
- The pi-cation interaction is preserved
- Disconnection----color shift

C

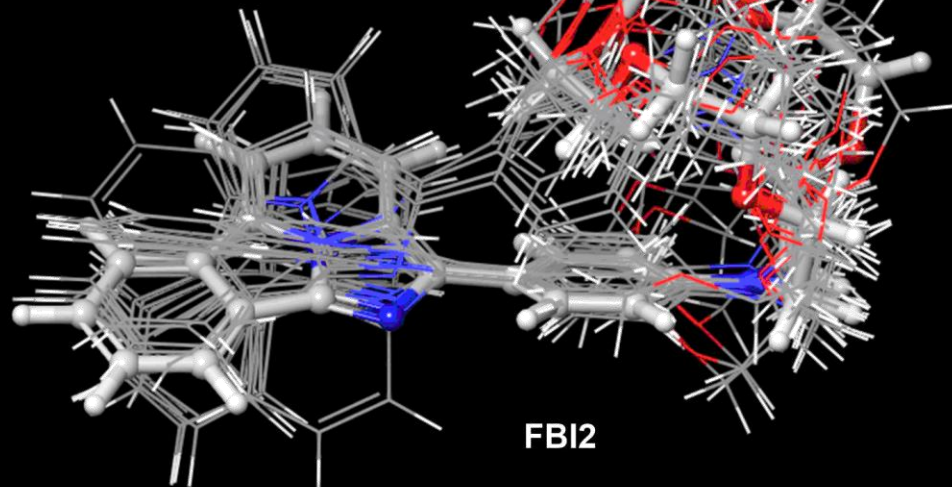
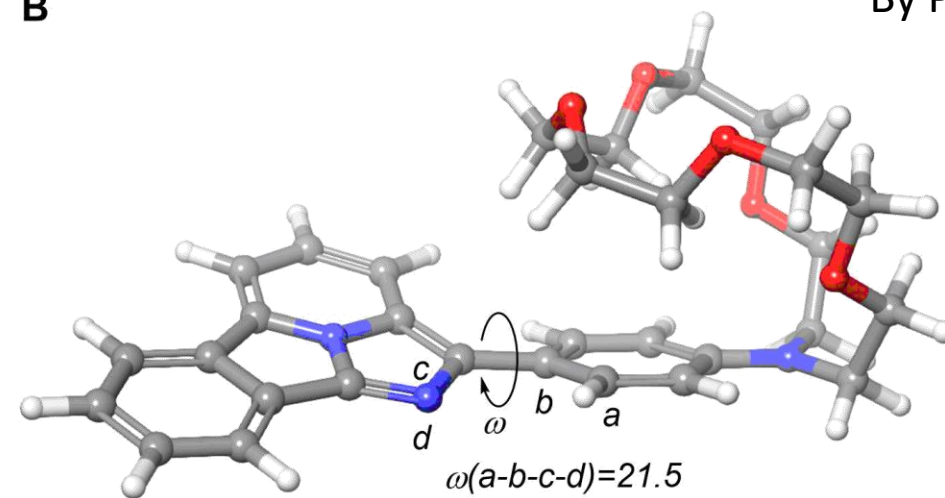
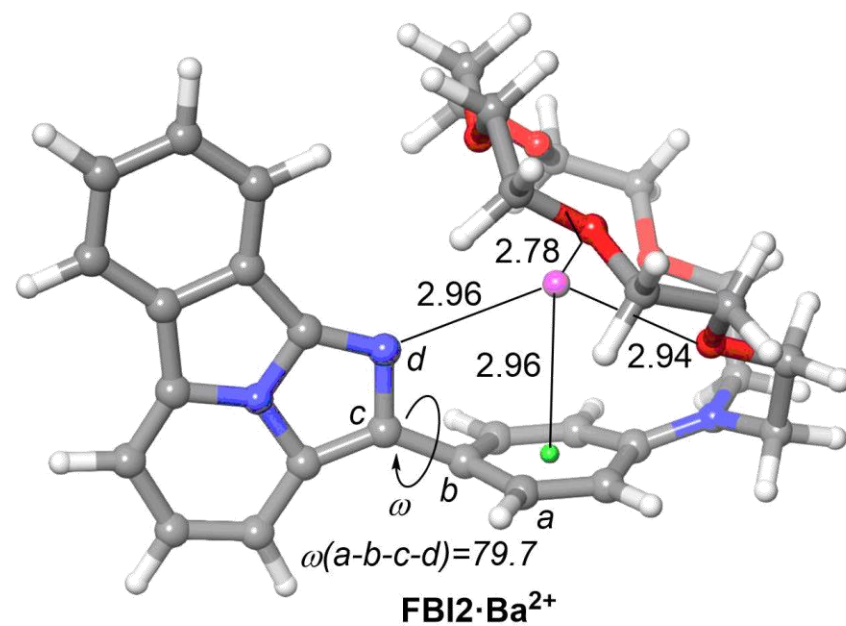
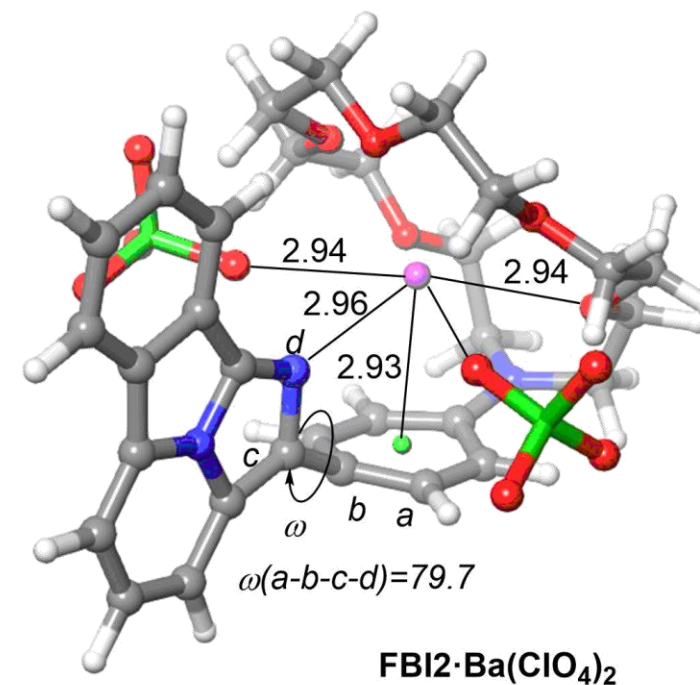
$$\Delta E_b(\text{FBI} - \text{G2}) = -172.5 \text{ kcal/mol}$$

$$\sum \Delta E_b(\text{optimal}) = -301.2 \text{ kcal/mol}$$

$$\Delta E(\text{deformation}) = +128.71 \text{ kcal/mol}$$

45

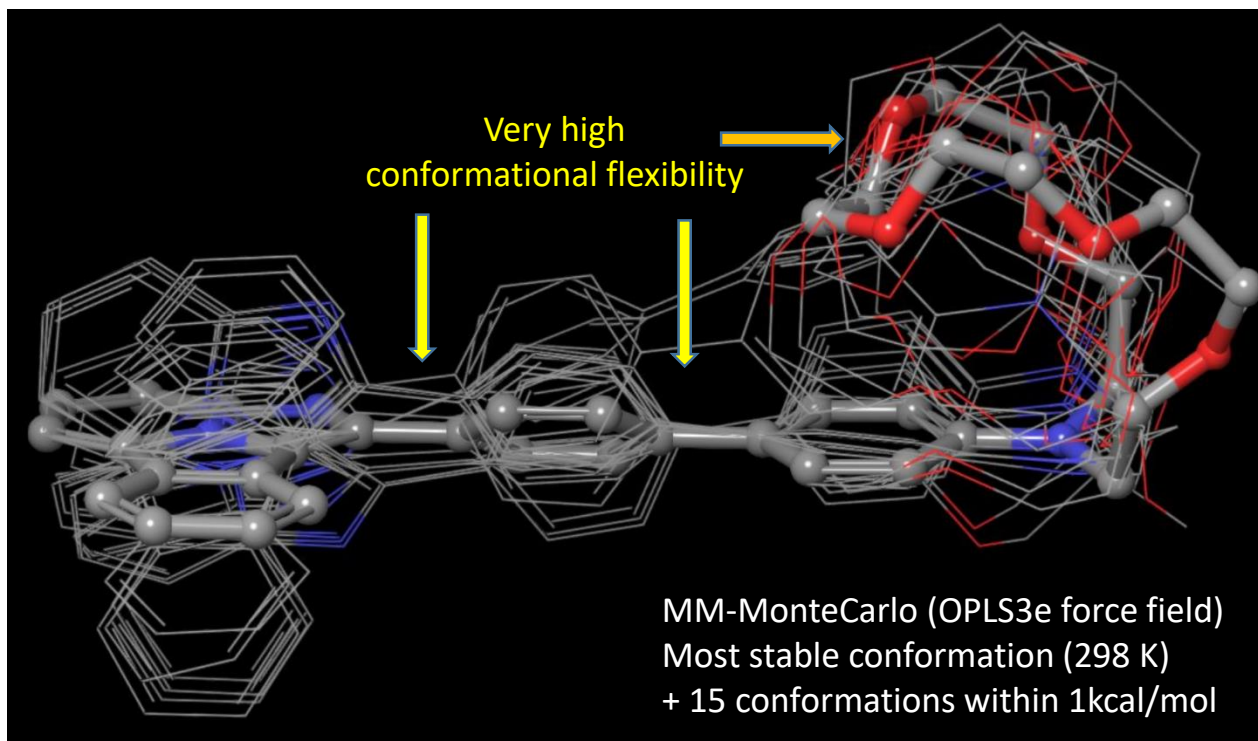
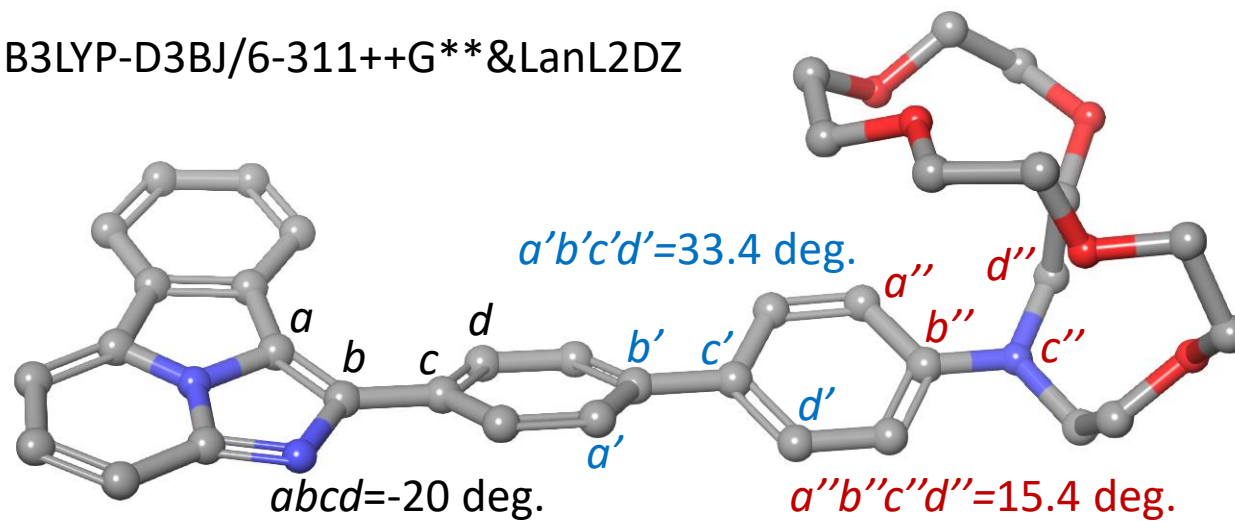
A Force field: OPLS3e
 $\Delta E(\text{MM})=4.0$ kJ/mol
 #Structures=18

**B****C****D**

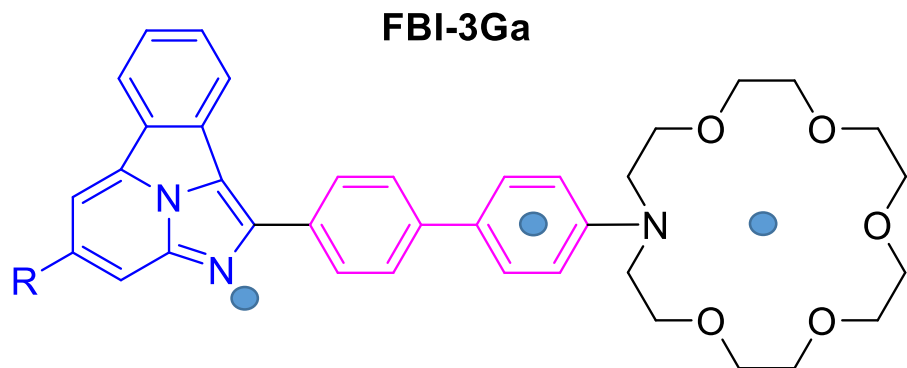
B3LYP-D3BJ/6 311++G(d,p)&LANL2DZ(Ba)

FBI-G3: The parent molecule FBI-G3a

B3LYP-D3BJ/6-311++G**&LanL2DZ



- Extended system
- High flexibility respect to G1
- Broad emission spectra

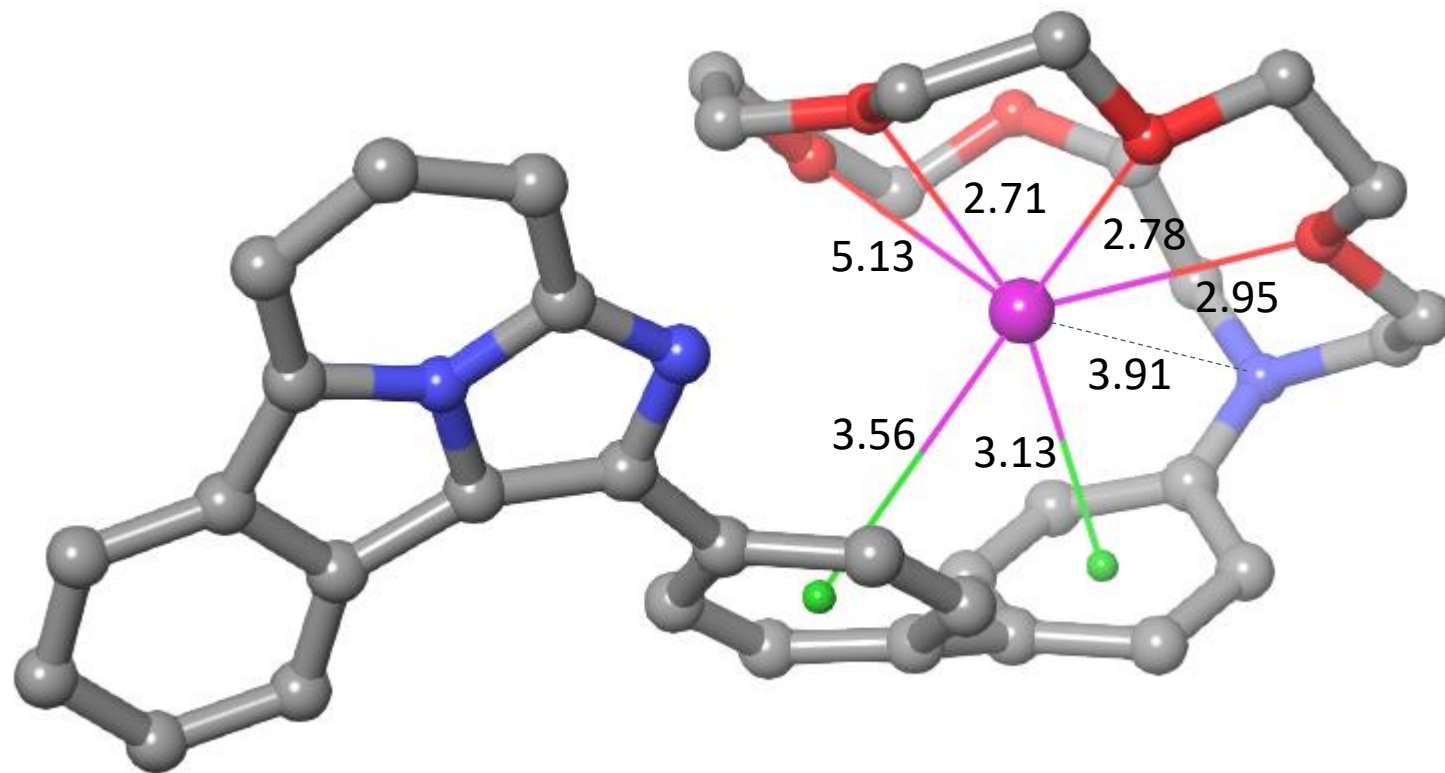


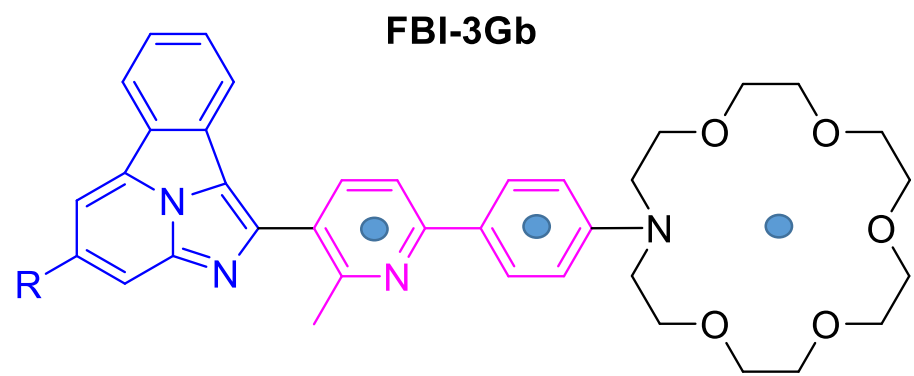
B3LYP-D3BJ/6-311++G**&LanL2DZ

$$\Delta E_b(\text{FBI} - \text{G3a}) = -157.4 \text{ kcal / mol}$$

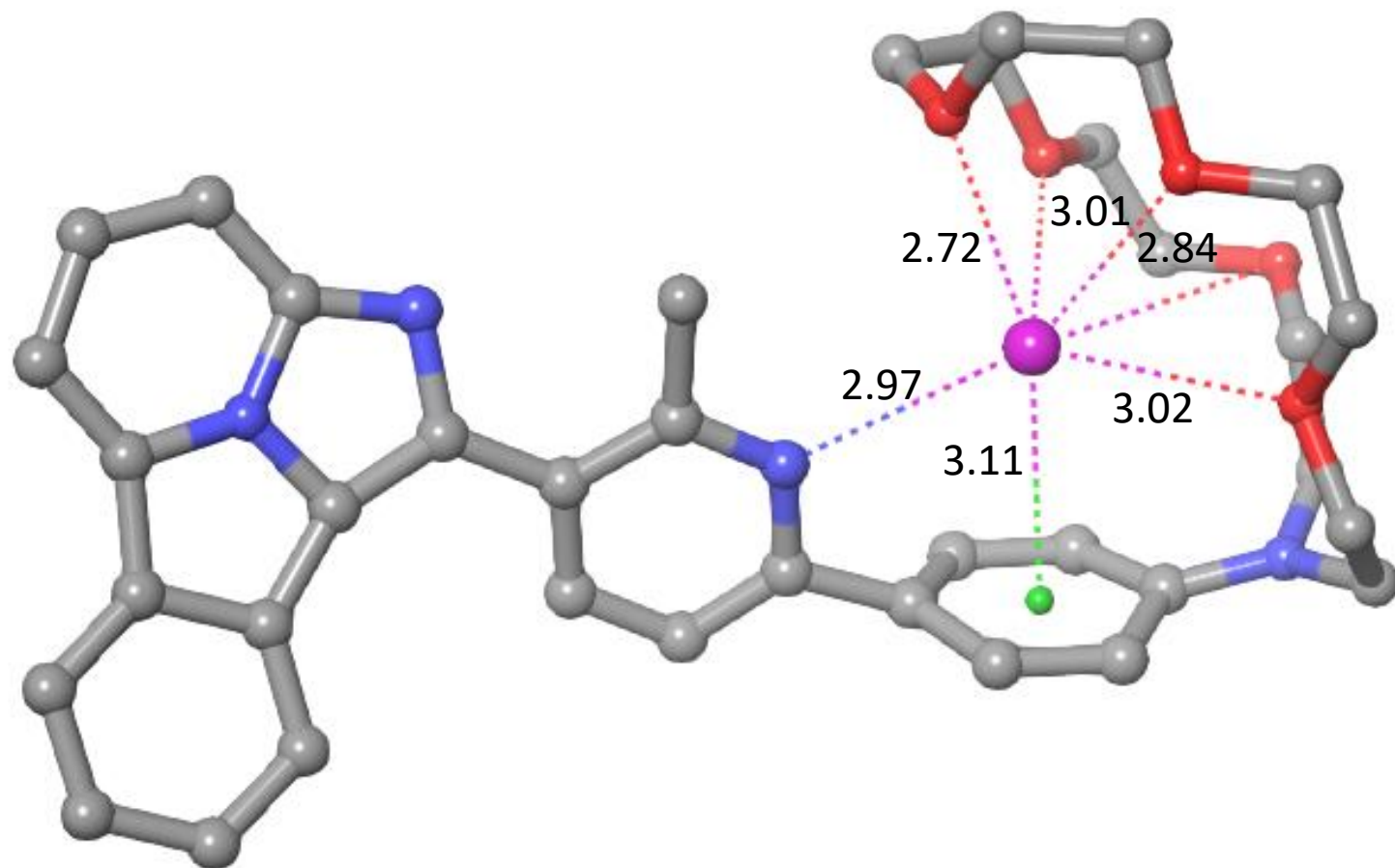
$$\sum \Delta E_b(\text{optimal}) = -315.7 \text{ kcal / mol}$$

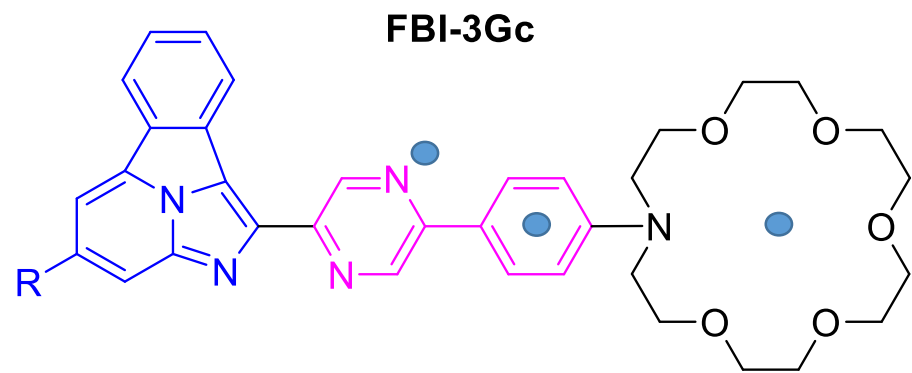
$$\Delta E(\text{deformation}) = +158.3 \text{ kcal / mol}$$



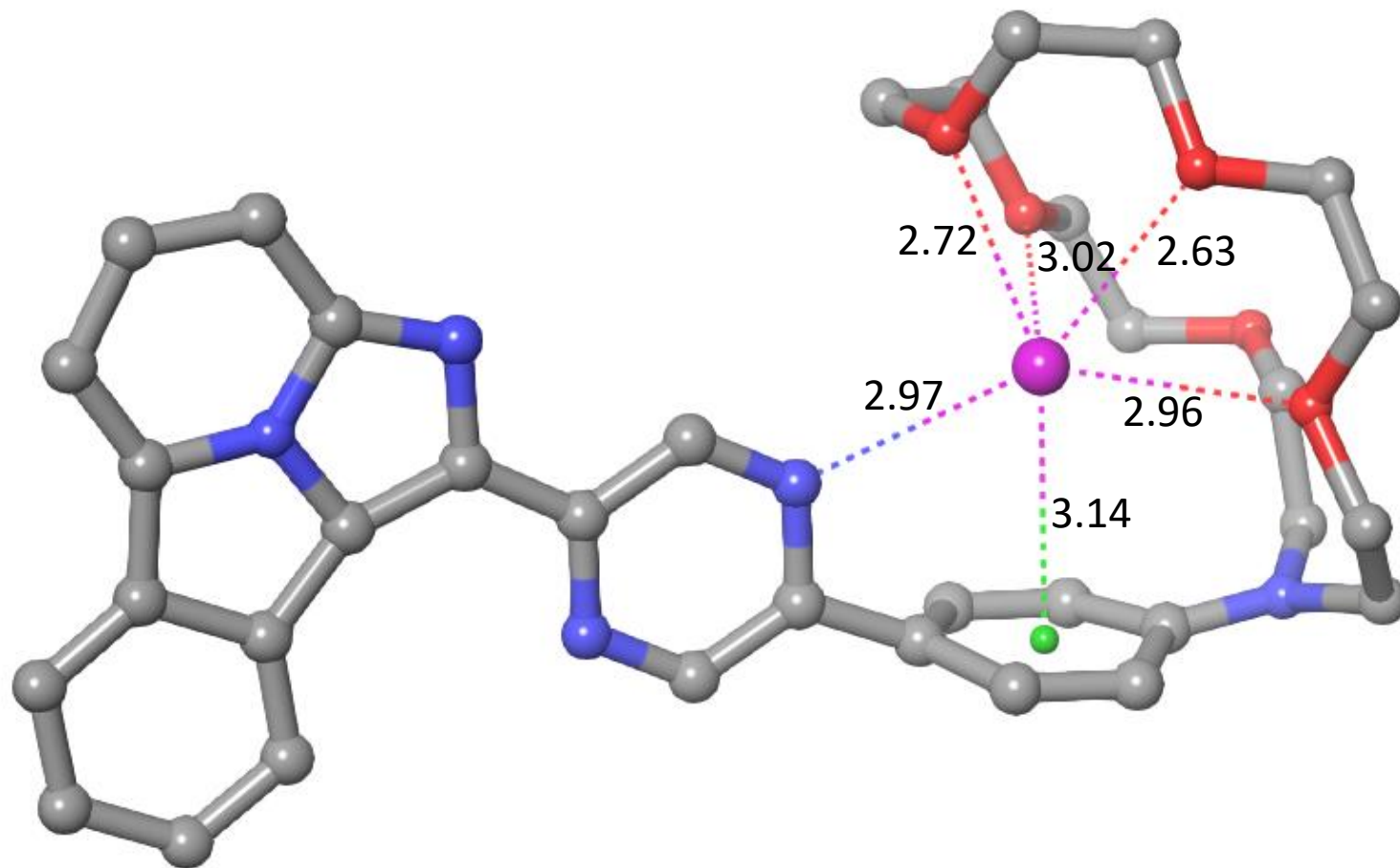


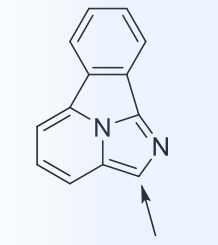
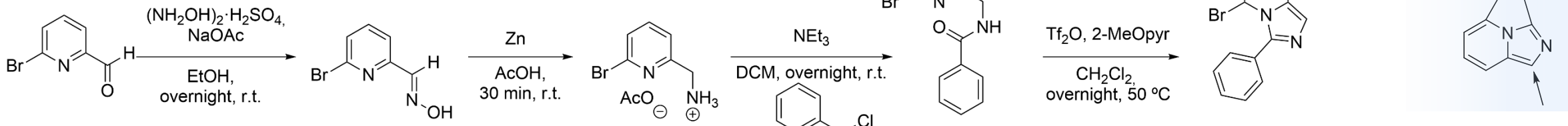
B3LYP-D3BJ/6-311++G**&LanL2DZ



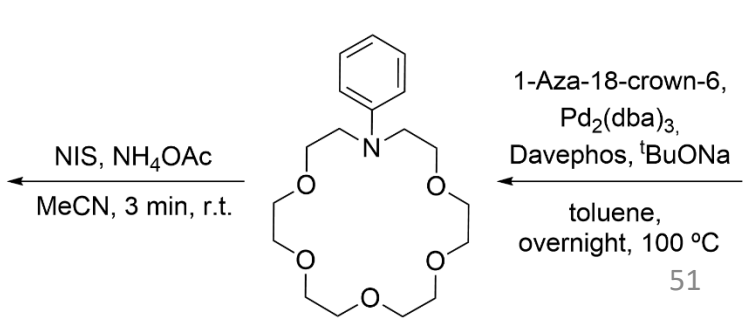
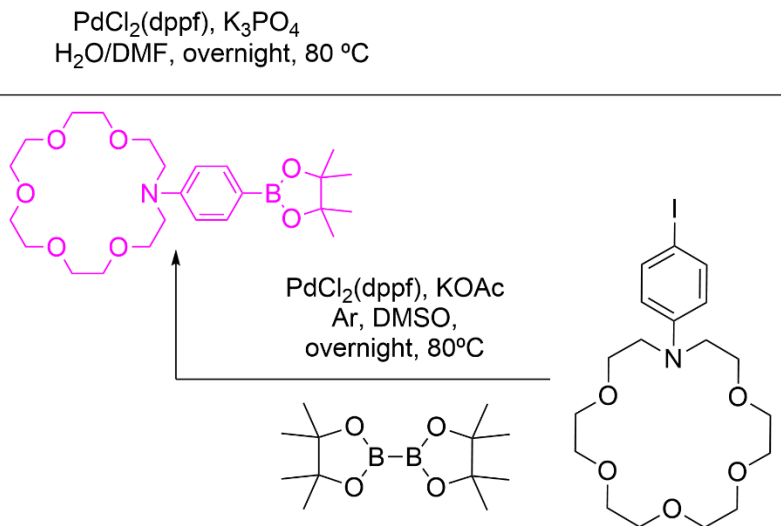
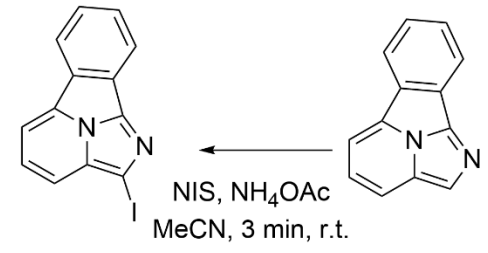
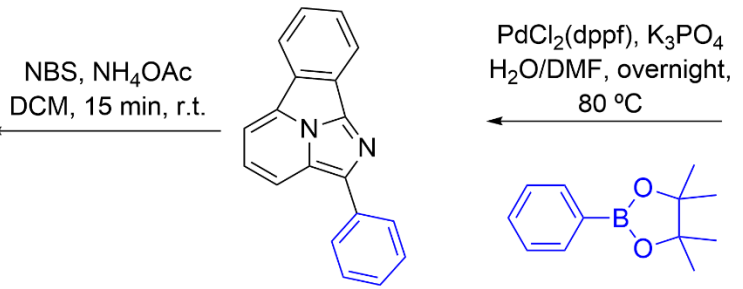
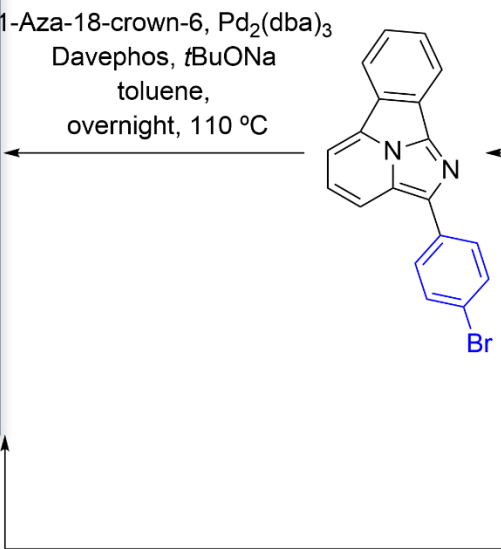
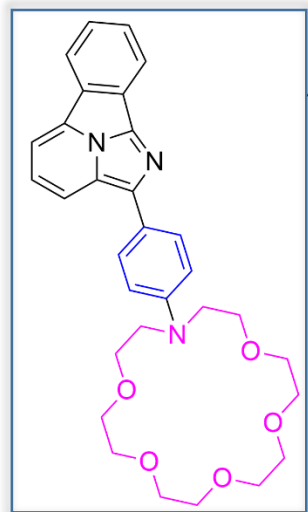


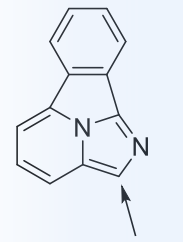
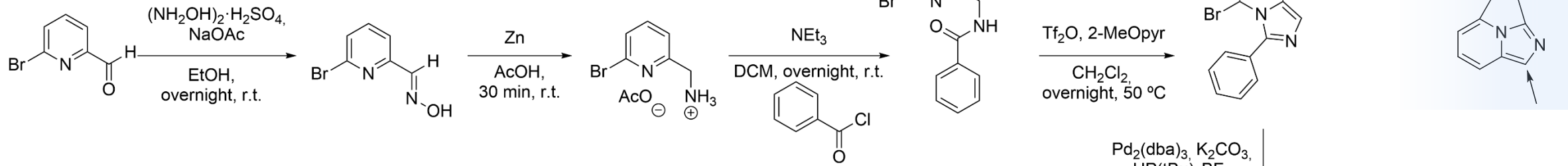
B3LYP-D3BJ/6-311++G**&LanL2DZ



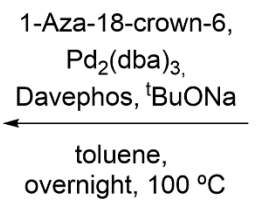
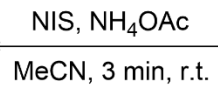
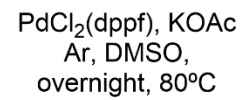
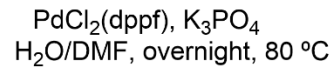
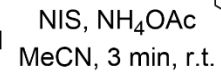
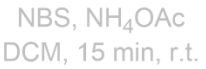
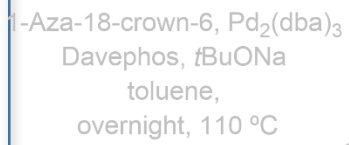
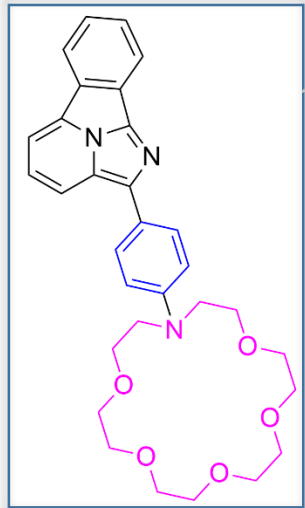


FBI-G2

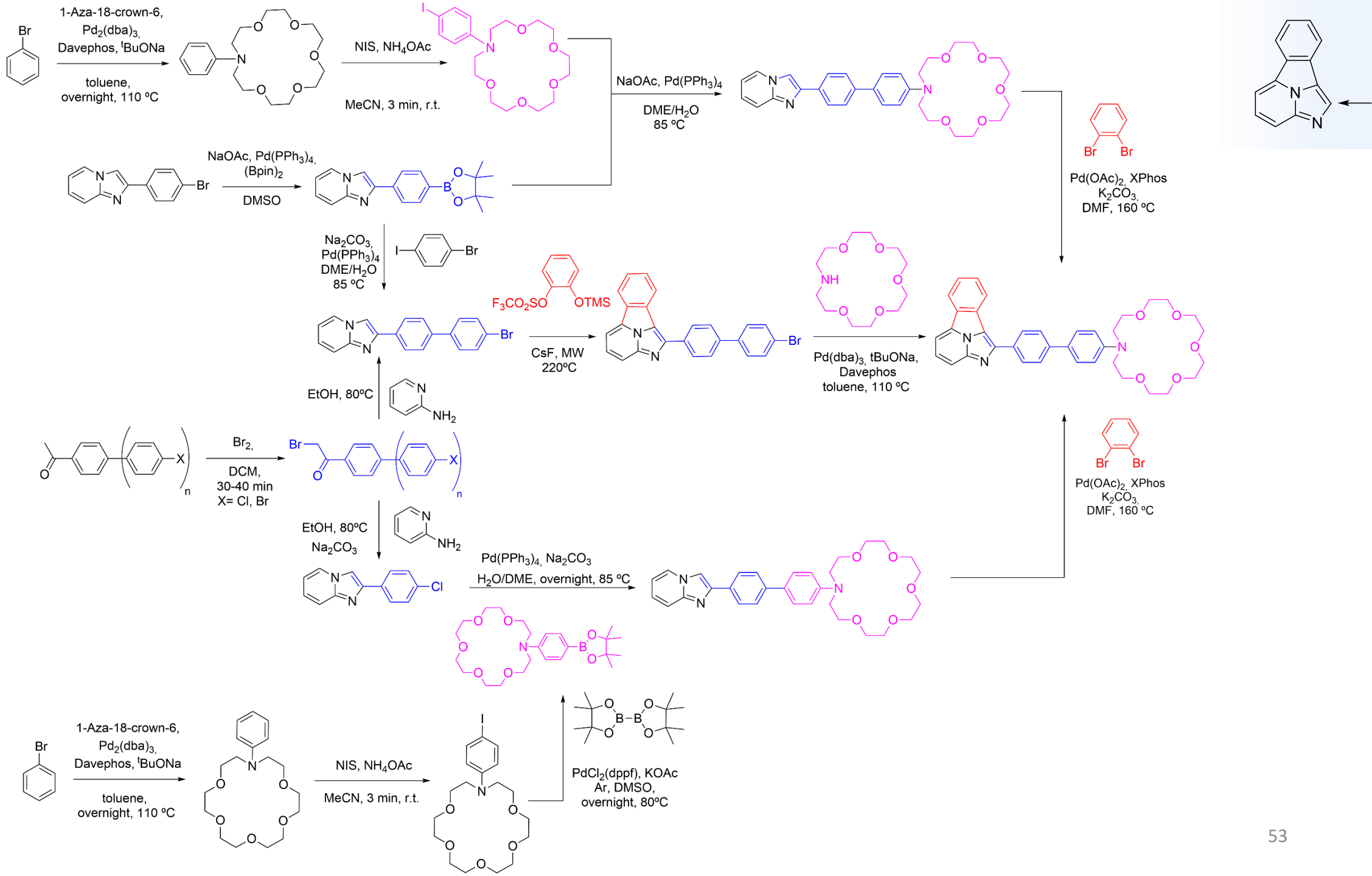




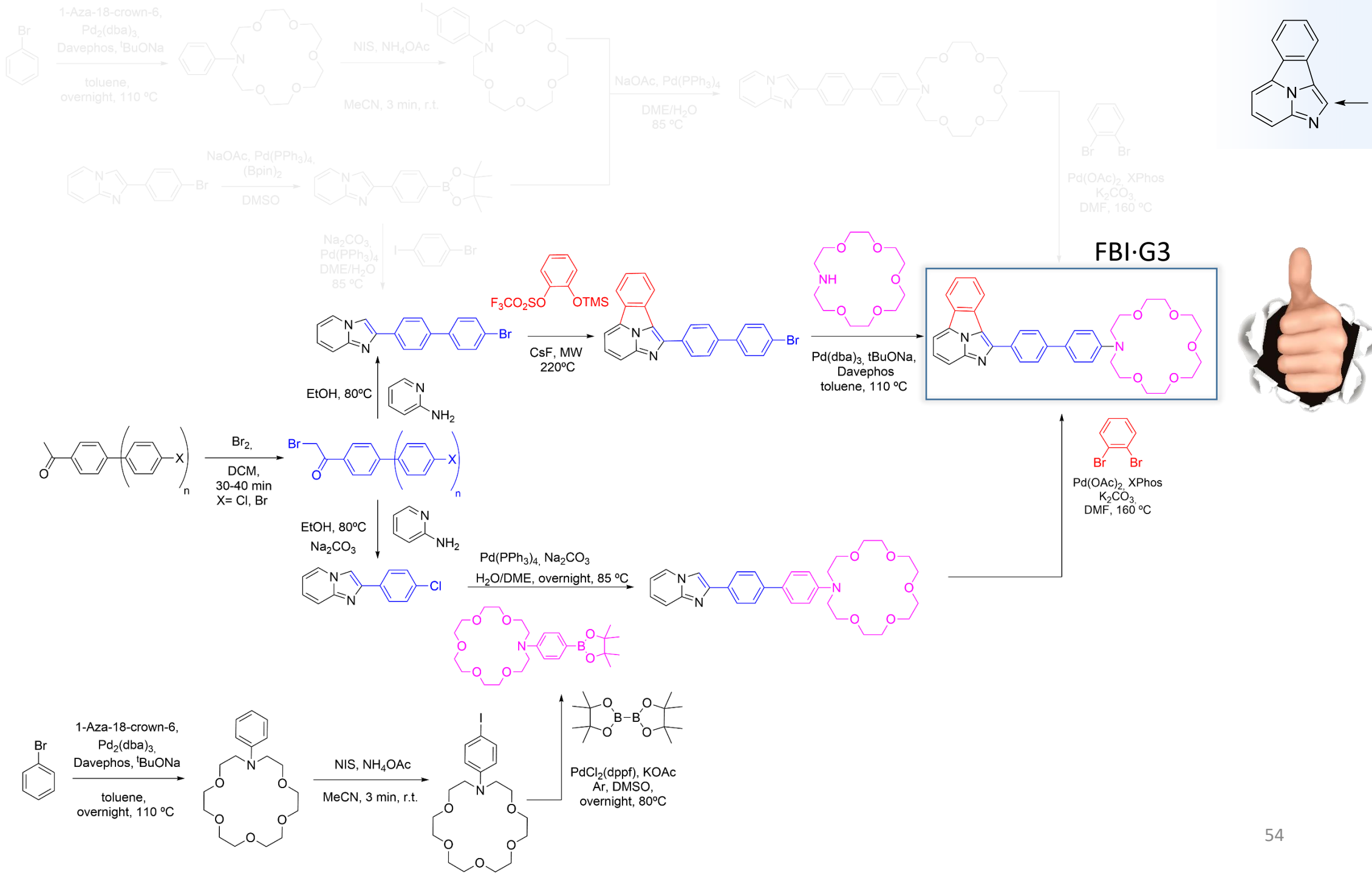
FBI-G2a



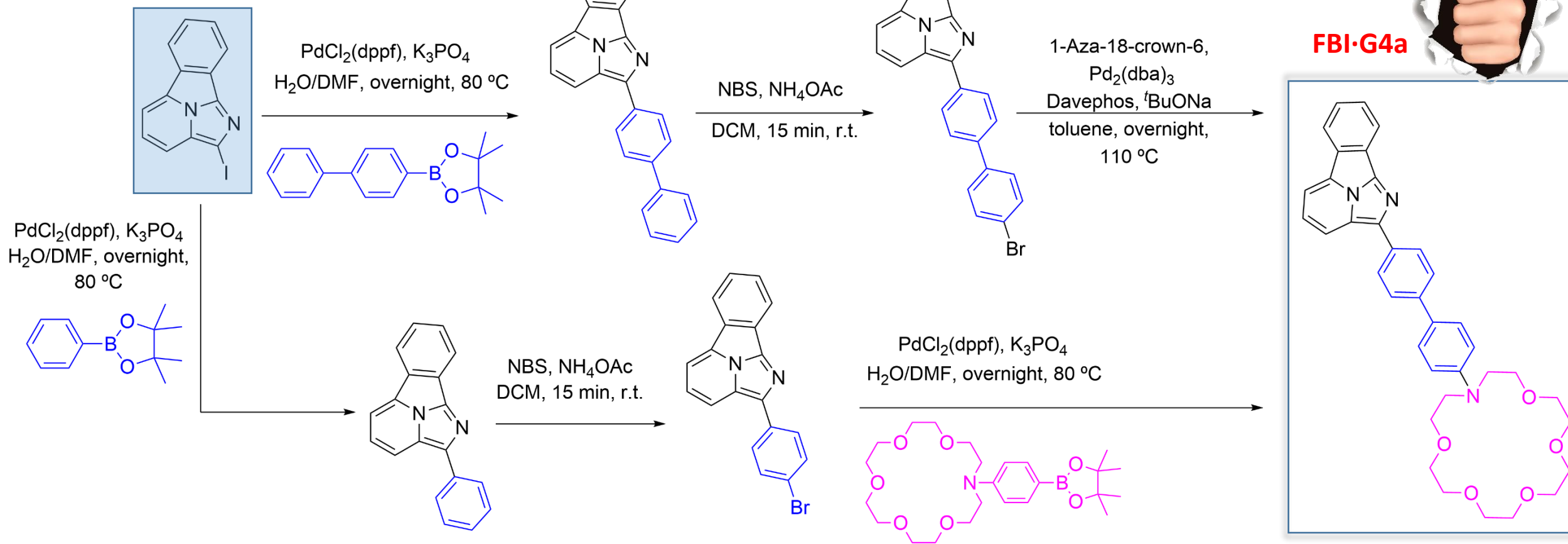
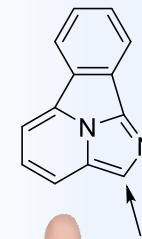
Chemical Synthesis of FBI-G3 Candidates



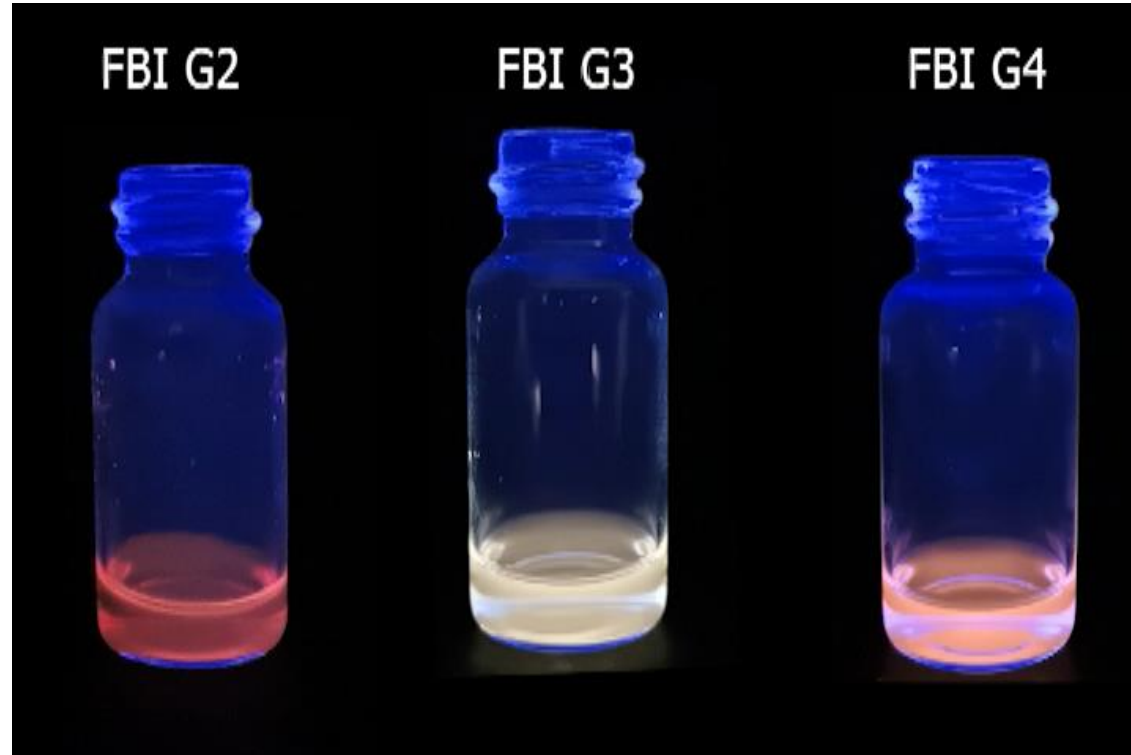
Chemical Synthesis of FBI-G3 Candidates



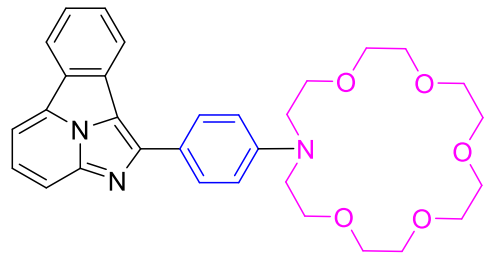
Chemical Synthesis of FBI-G4 Candidates



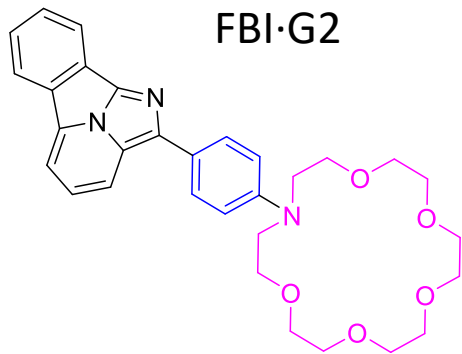
Fluorescent Bicolor Indicators G2, G3 and G4



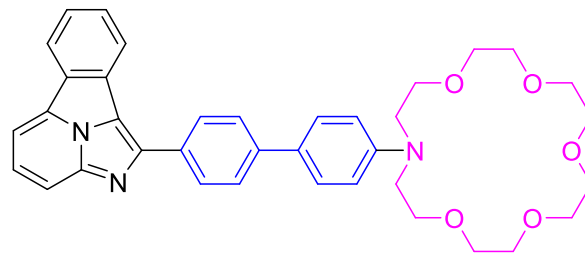
FBI-G1



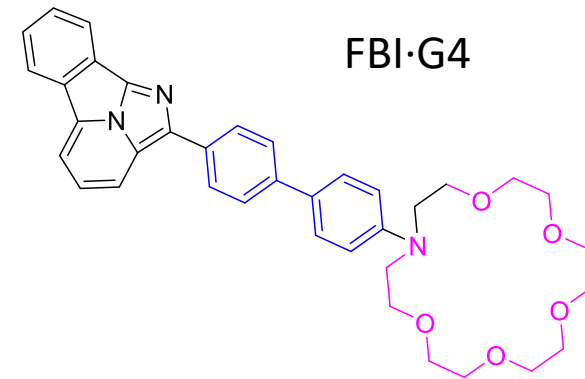
FBI-G2



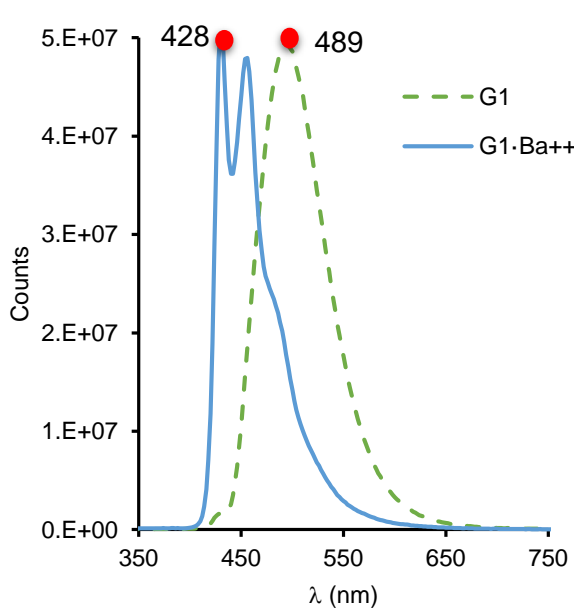
FBI-G3



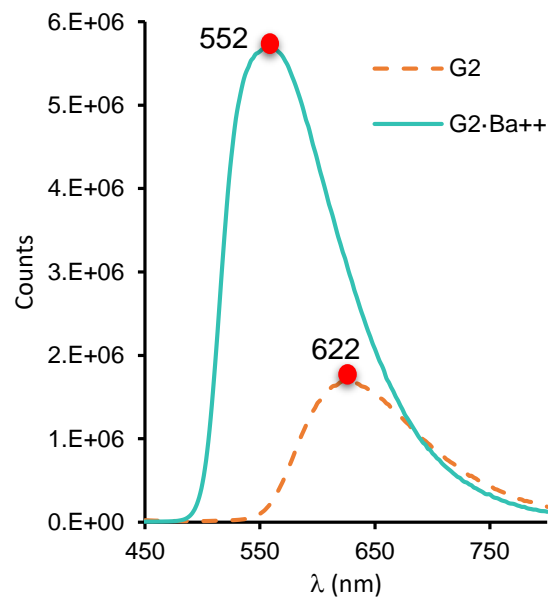
FBI-G4



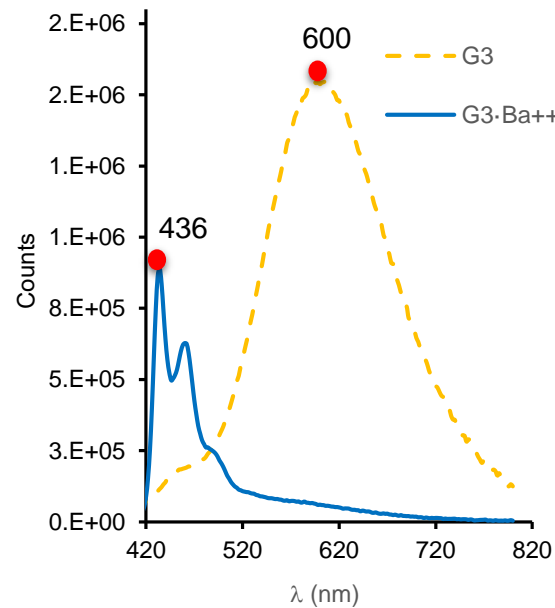
G1 vs G1·Ba²⁺



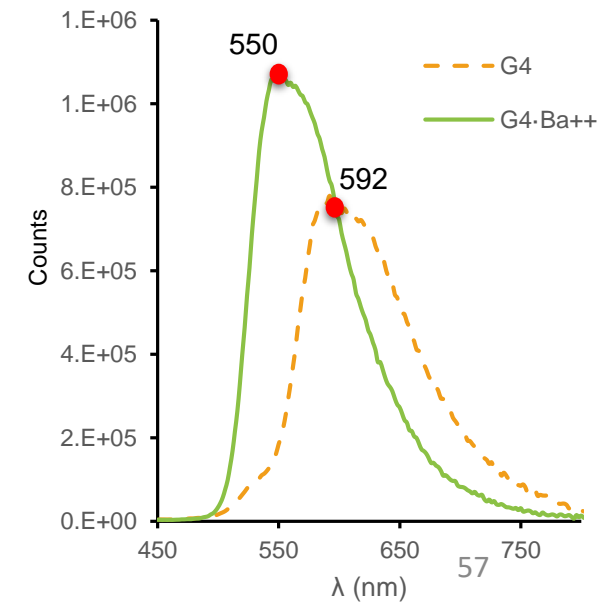
G2 vs G2·Ba²⁺

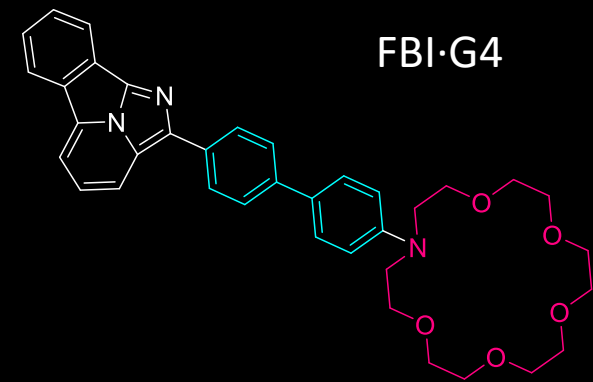
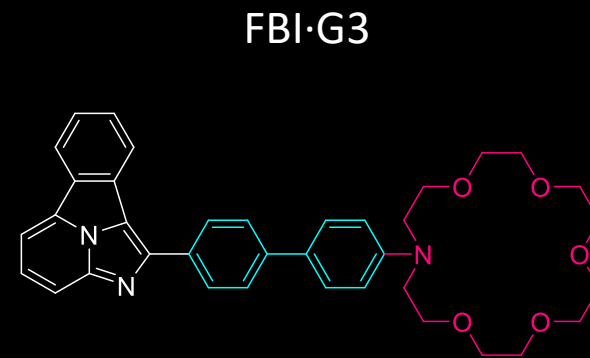
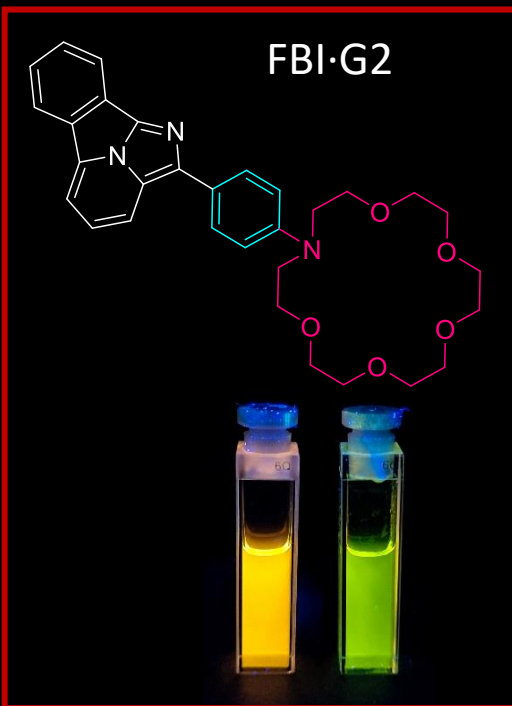
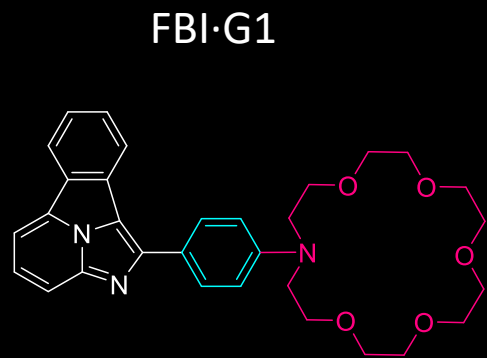


G3 vs G3·Ba²⁺



G4 vs G4 Ba²⁺

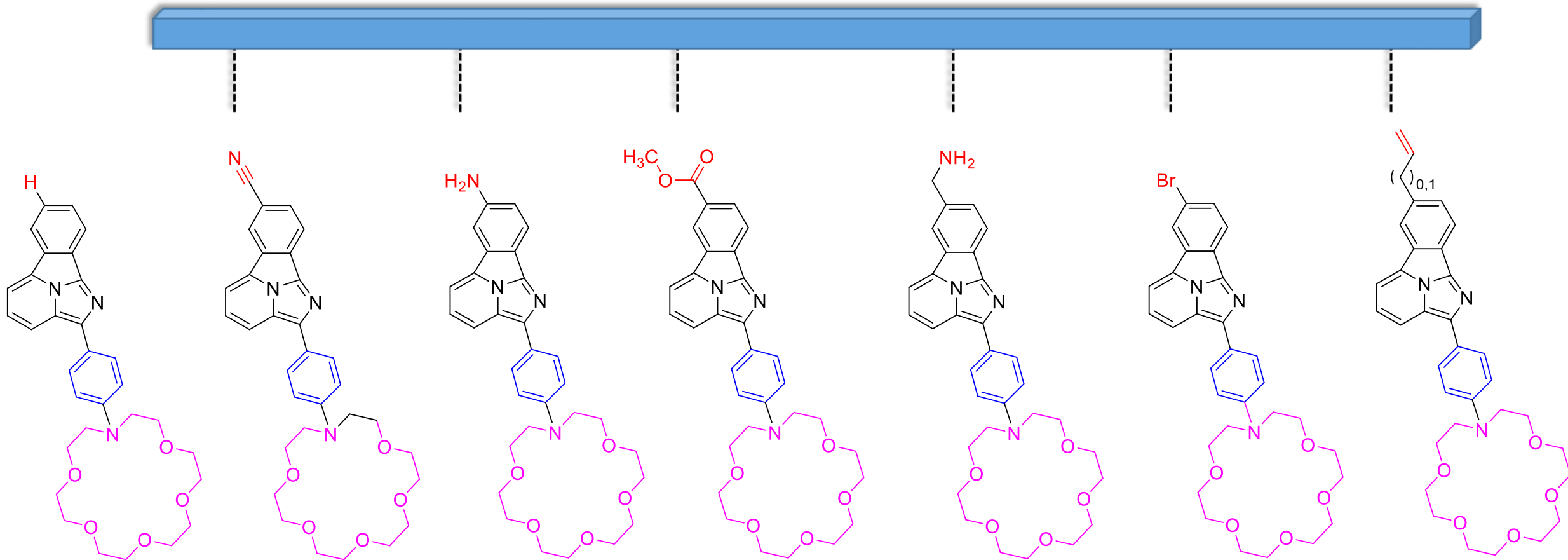




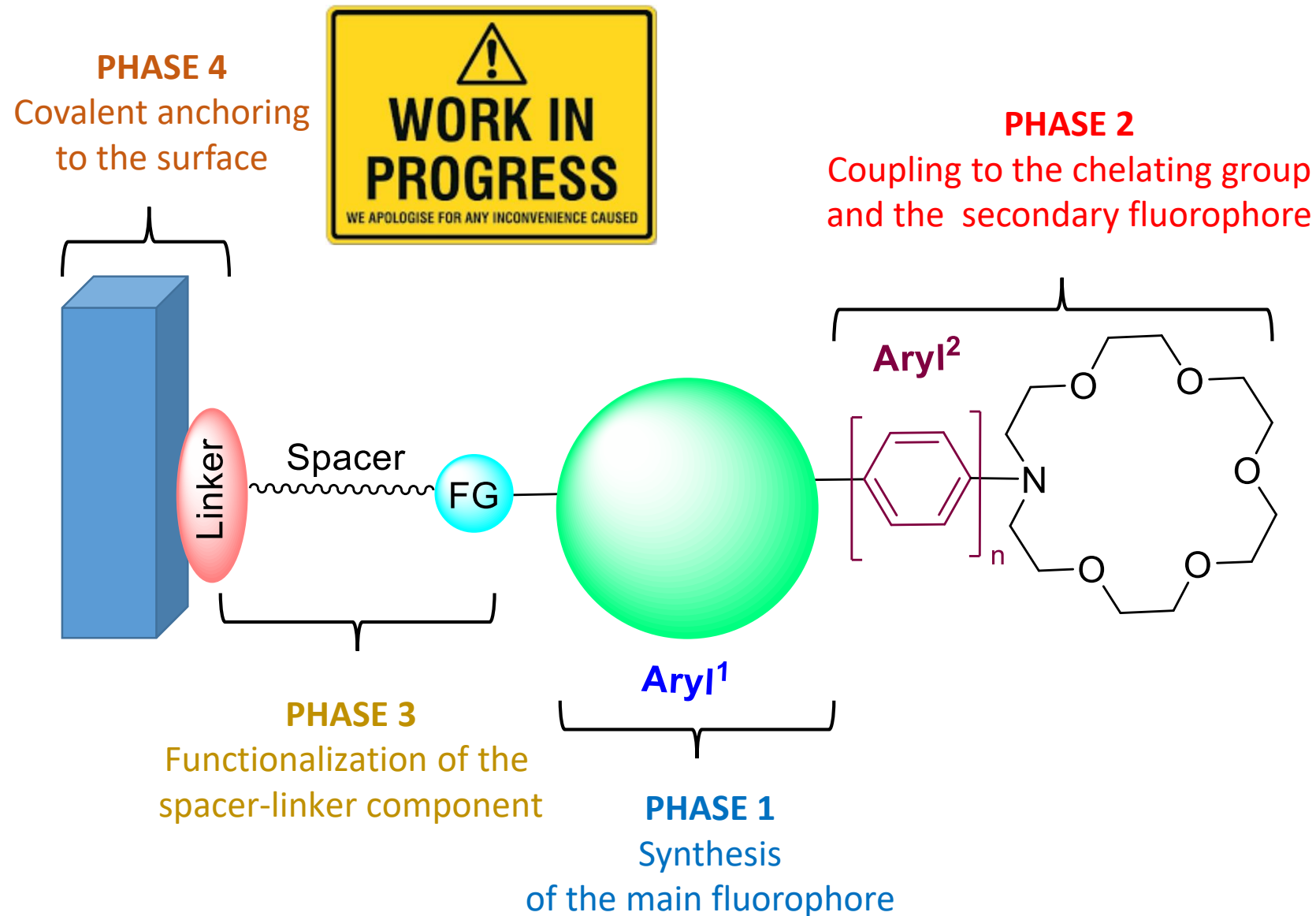
Compound	λ_{em}^a (nm)		$\Delta\lambda$ (nm)	$f_{\lambda_{em}}^b$	ϕ_{λ}^c		B_{λ}^c ($M^{-1} cm^{-1}$)		ϵ_{λ}^c ($M^{-1} \cdot cm^{-1}$)		σ_{λ}^c (\AA^2)		K_a^d (M^{-1})
	Gn	Gn·Ba ²⁺			Gn	Gn·Ba ²⁺	Gn	Gn·Ba ²⁺	Gn	Gn·Ba ²⁺	Gn	Gn·Ba ²⁺	
G1	489	428	61	179.7	0.67	0.45	16,388	9,937	24,460	22,084	0.94	0.84	$5.3 \cdot 10^4$
G2	622	552	70	210.6	0.23	0.55	5,990	16,564	26,044	30,117	1.00	1.15	$1.3 \cdot 10^6$
G3	600	436	164	5.5	0.74	0.61	13,380	3,068	18,081	5,031	0.69	0.19	$5.0 \cdot 10^5$
G4	592	550	42	8.2	0.41	0.58	5,193	5,794	12,667	9,991	0.48	0.38	$1.0 \cdot 10^5$

^a Emission wavelengths λ_{em} at a $\lambda_{exc-max}$. ^b Peak discrimination factors (f_{λ}) with respect to unbound fluorophores Gn at λ_{em} . ^c All values for Quantum yields ϕ_{λ} , molecular brightness of the fluorescent emissions (B_{λ}), molar extinction coefficient and cross section were recorded for $\lambda(G1) = 250$ nm; $\lambda(G2) = 325$ nm; $\lambda(G3) = 368$ nm; $\lambda(G4) = 354$ nm. ^d K_a with barium perchlorate in acetonitrile at 298 K using the Benesi–Hildebrand method.

Differently Functionalized FBI-G2 compounds

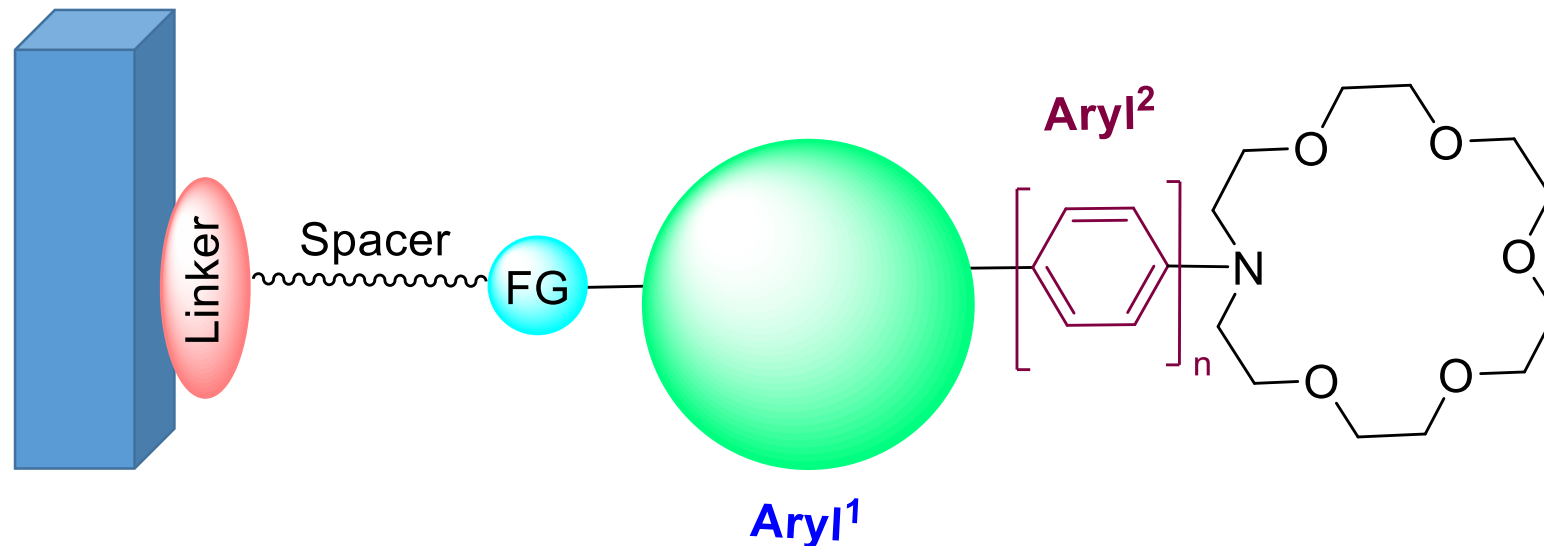


A unified synthetic scheme towards functional surfaces



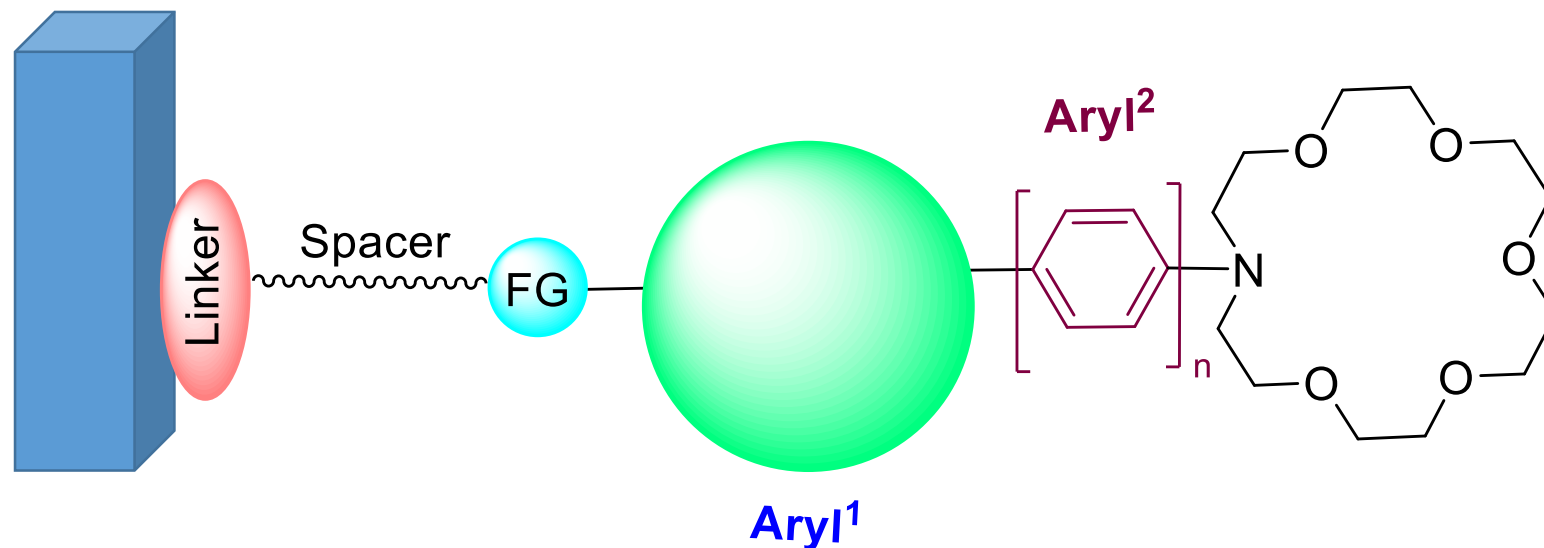
Conclusions and Outlook

- It is possible to expand and optimize the configurational space of fluorescent bicolor indicators (FBIs) by modifying the structure of the heteroaromatic polycyclic ($Aryl^1$) and the *para*-phenylene/biphenylene ($Aryl^2$) components.



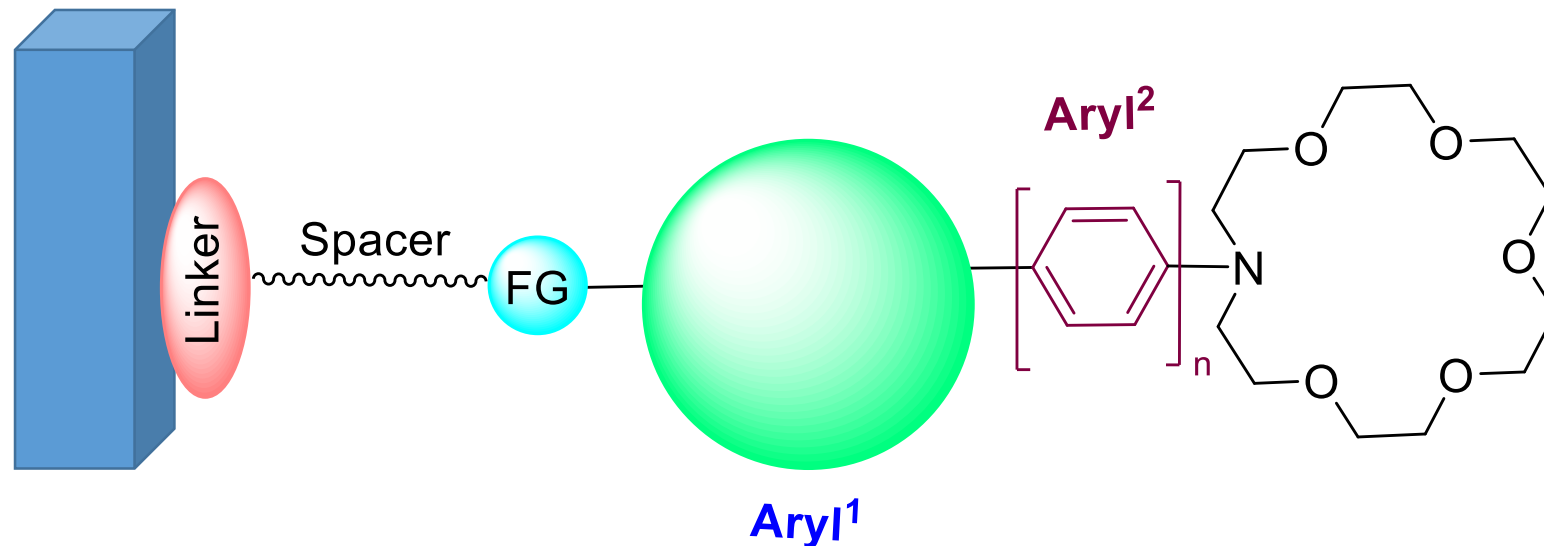
Conclusions and Outlook

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- Among the different options synthesised and tested, the FBI-G2 series is particularly promising.



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- It is possible to expand and optimize the configurational space of fluorescent bicolor indicators (FBIs) by modifying the structure of the heteroaromatic polycyclic ($Aryl^1$) and the *para*-phenylene/biphenylene ($Aryl^2$) components.
- Among the different options synthesised and tested, the FBI-G2 series is particularly promising.
- Work is in progress to modify this novel family of sensors in order to generate suitable functionalized surfaces.





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misaotra
nandri

Searching the Grail: A background free $\beta\beta 0\nu$ experiment using Ba^{2+} tagging in a High-Pressure Xenon Chamber

Iván Rivilla and on behalf of 

Zoraida Freixa, Fernando Auria-Luna, Juan Molina-Canteras, Mikel Odriozola-Gimeno, Borja Aparicio, Ane I. Aranburu, Amaia Larumbe, Virginia San Nacienceno, Nerea Alberro, Pablo García, Francesc Monrabal, Pablo Herrero, Celia Rogero, Juan J. Gómez-Cadenas and Fernando P. Cossío

