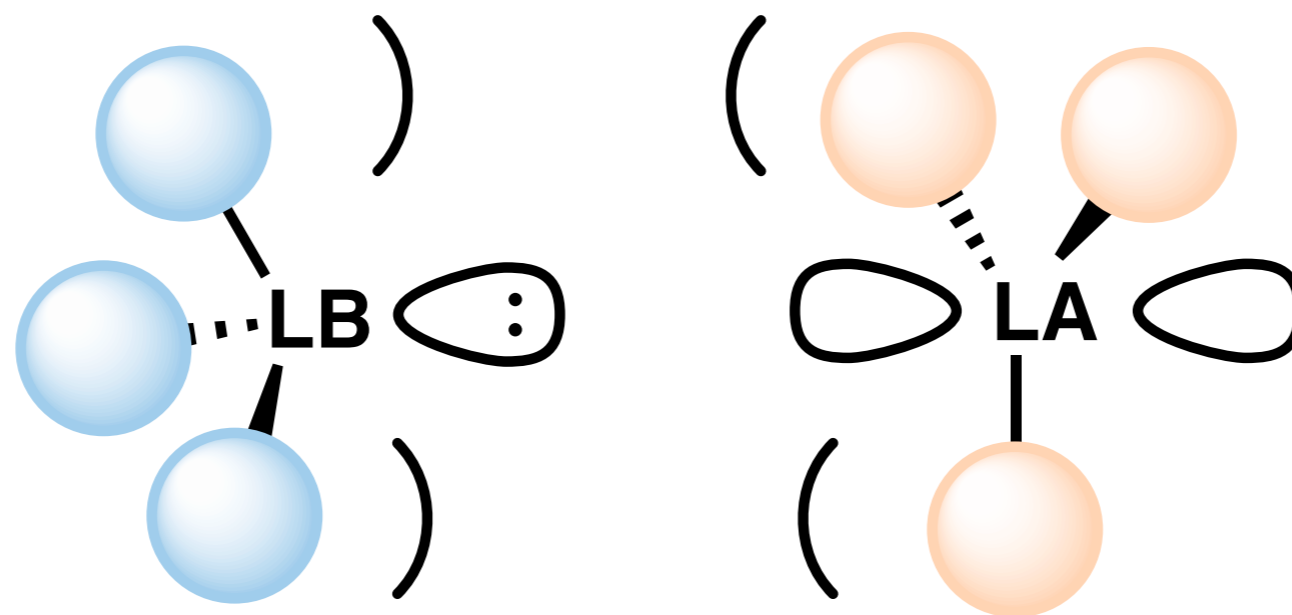
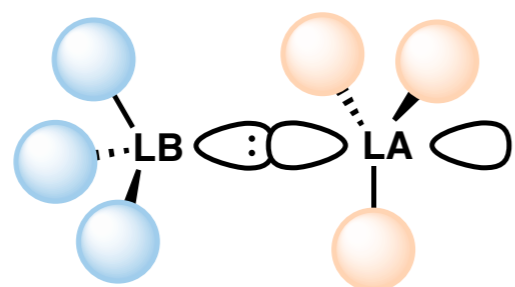
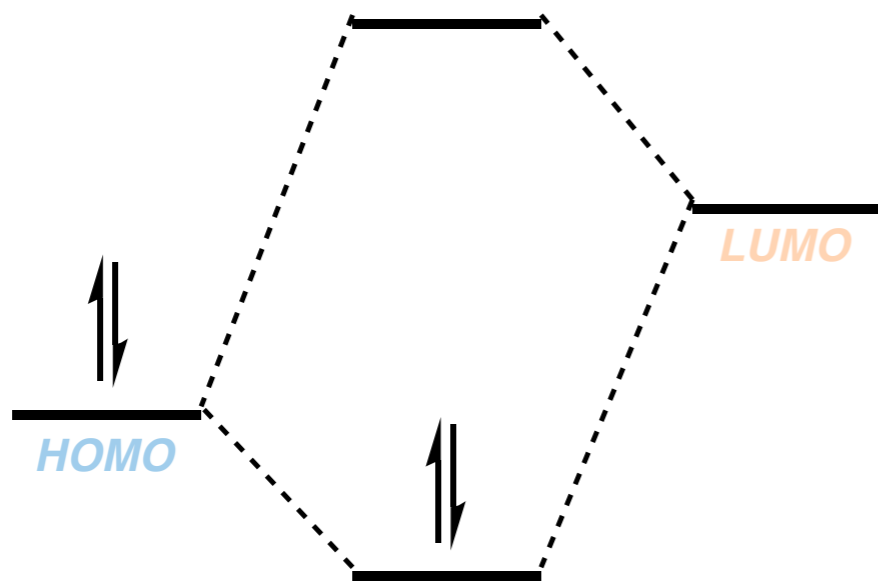


Frustrated Lewis Pairs

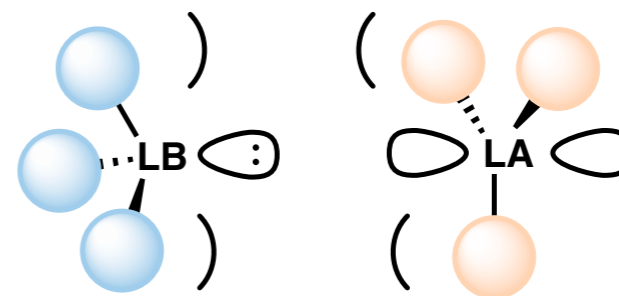
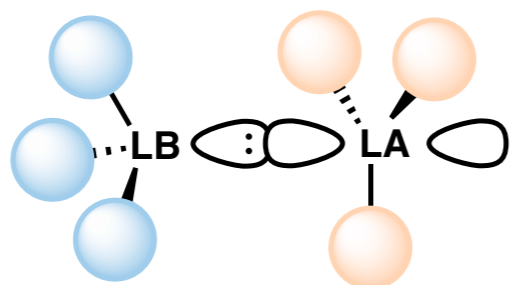
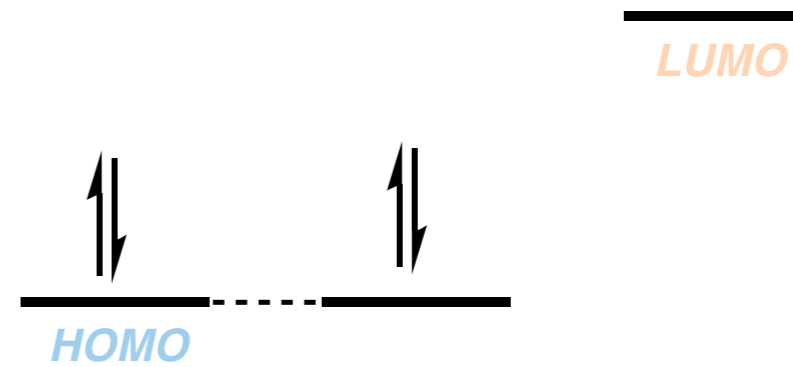
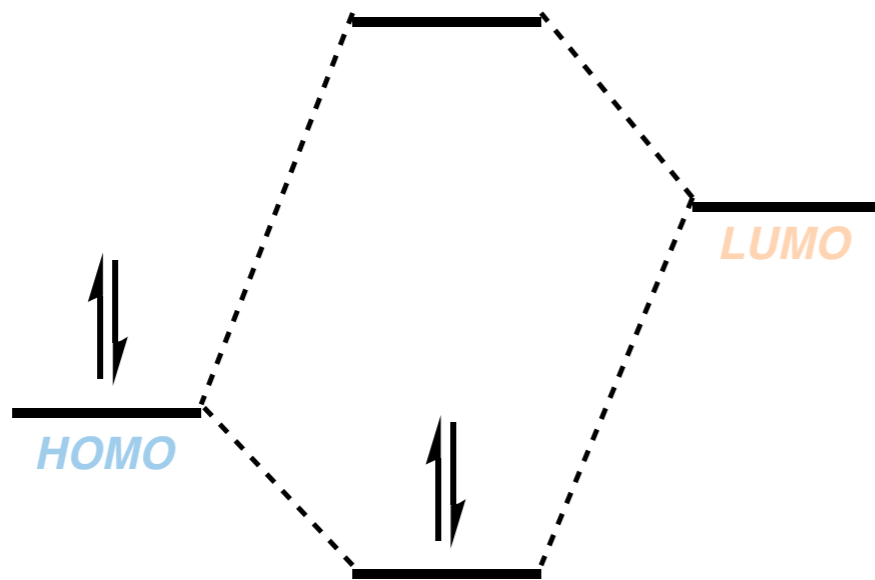


Johanna M. Masterson
Literature Meeting
Sorensen Group Meeting
June 25, 2021

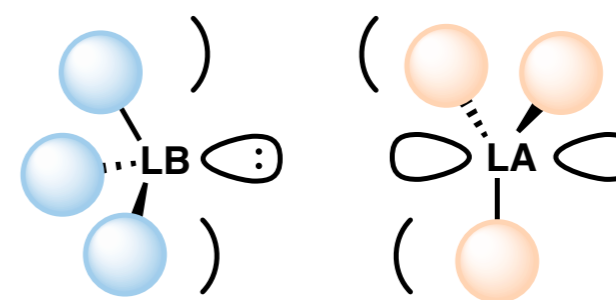
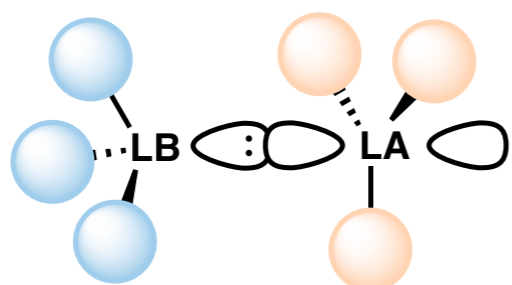
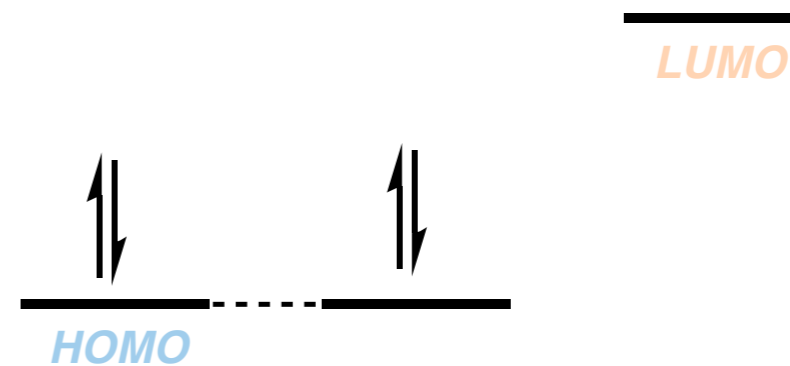
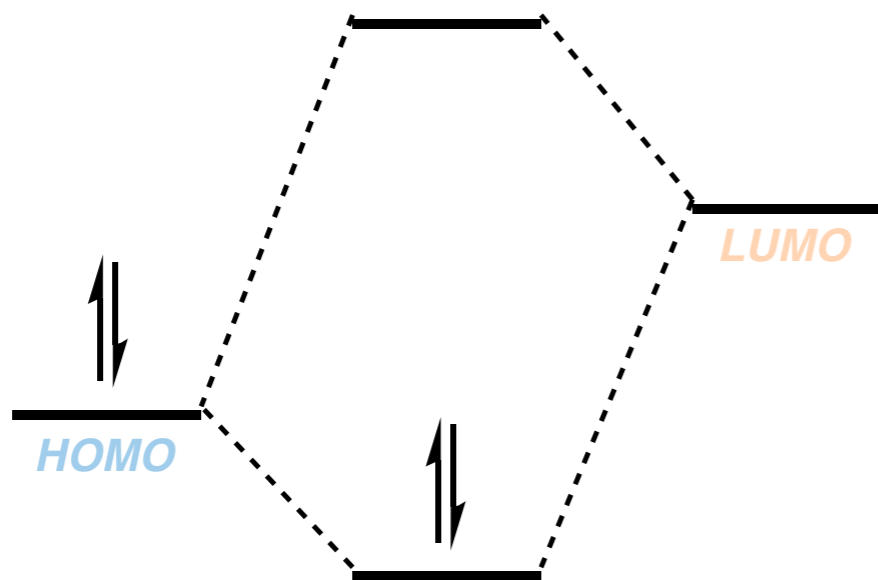
Introduction to FLPs



Introduction to FLPs

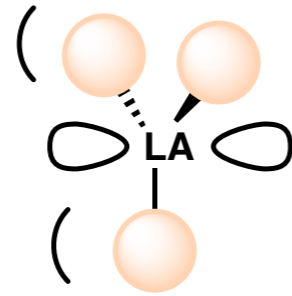
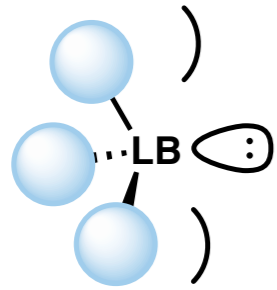


Introduction to FLPs

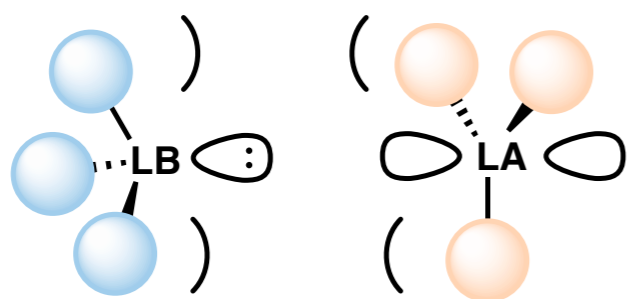
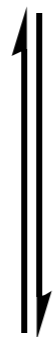


*steric/geometric constraints
dissociative equilibrium*

Mechanism and General Reactivity of FLPs

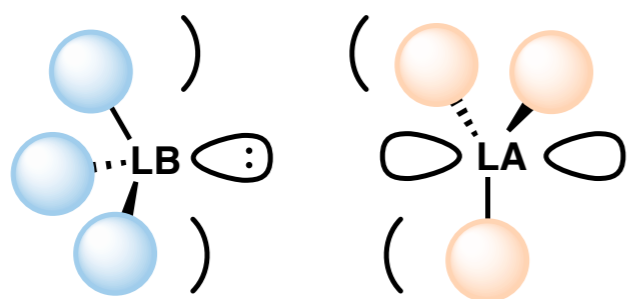
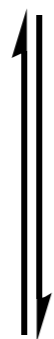


Mechanism and General Reactivity of FLPs



encounter complex

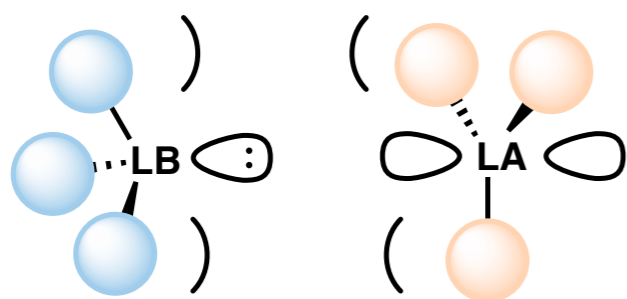
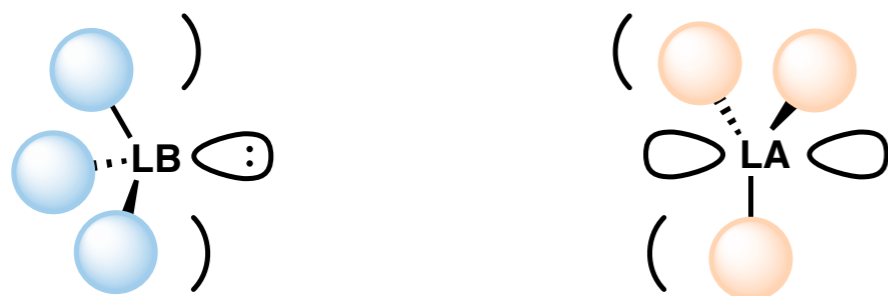
Mechanism and General Reactivity of FLPs



encounter complex

- **DFT and MD Studies**
(aggregates energetically favored)

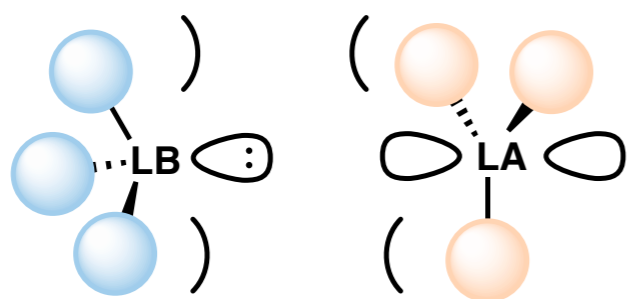
Mechanism and General Reactivity of FLPs



encounter complex

- **DFT and MD Studies**
(aggregates energetically favored)
- **NOE Experiments**
(directionless association driven by dispersion effects, slightly endergonic)

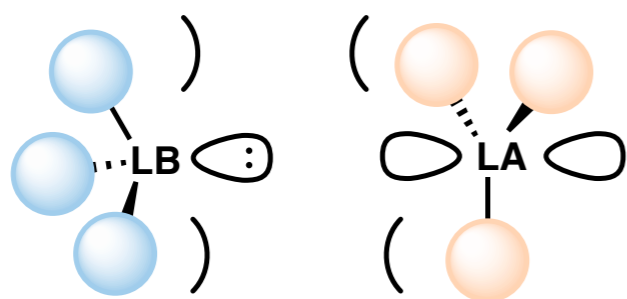
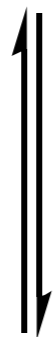
Mechanism and General Reactivity of FLPs



encounter complex

- **DFT and MD Studies**
(aggregates energetically favored)
- **NOE Experiments**
(directionless association driven by dispersion effects, slightly endergonic)
- **Neutron scattering/NMR in ionic liquid** (5% in solution)

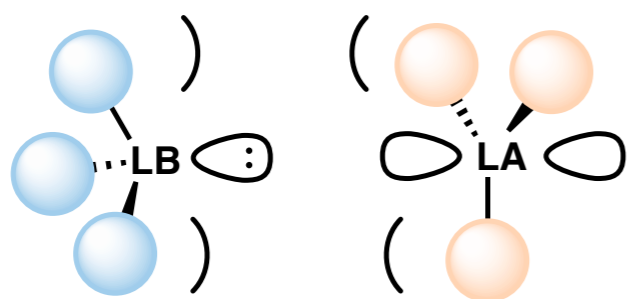
Mechanism and General Reactivity of FLPs



encounter complex

- **Cumulative acidity and basicity matter** (weaker acid requires stronger base and vice versa)

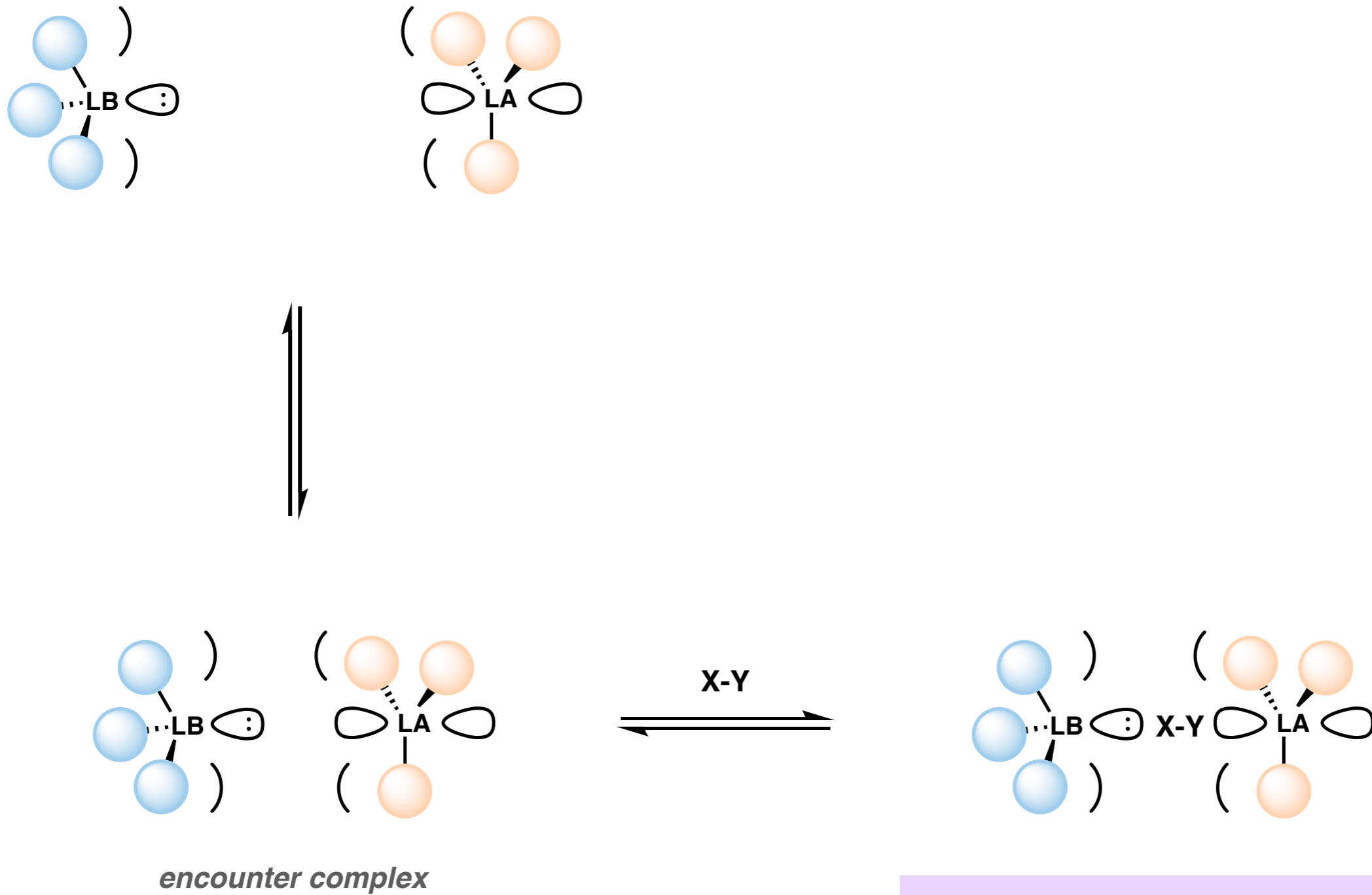
Mechanism and General Reactivity of FLPs



encounter complex

- **Cumulative acidity and basicity matter** (weaker acid requires stronger base and vice versa)
- **Distance between LA/LB centers** (3-5 angstroms is Goldilocks distance required for reactivity)

Mechanism and General Reactivity of FLPs

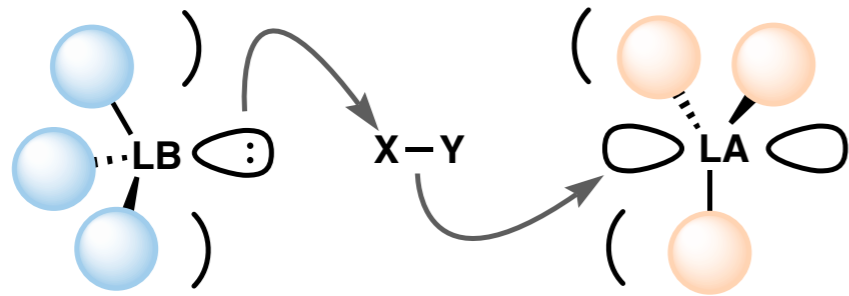


encounter complex

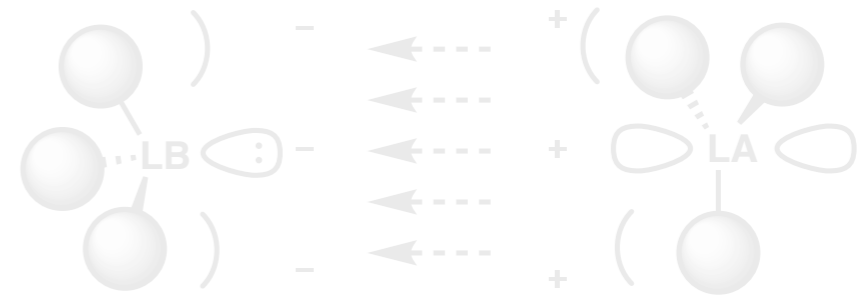
Two models for bond activation



Introduction to FLPs



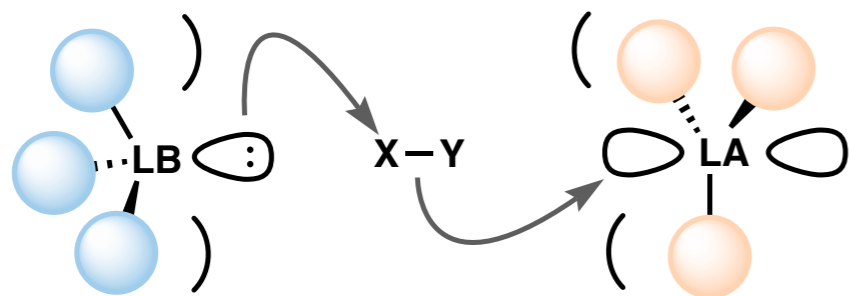
Electron Transfer Model



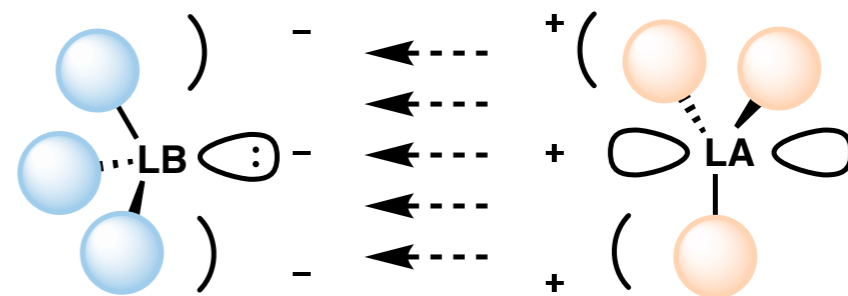
Electric Field Model

Distortion required for orbital overlap

Introduction to FLPs



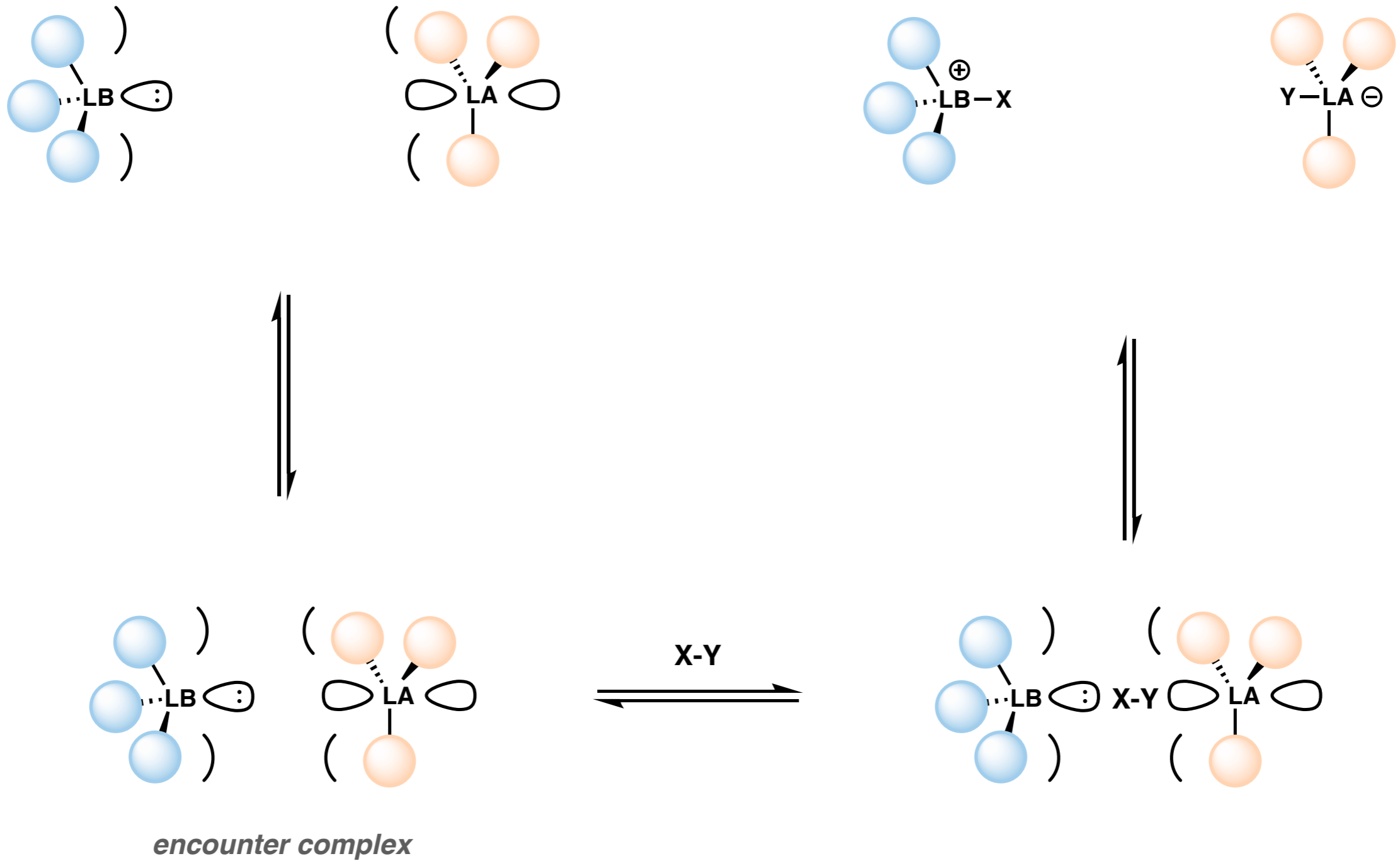
Electron Transfer Model



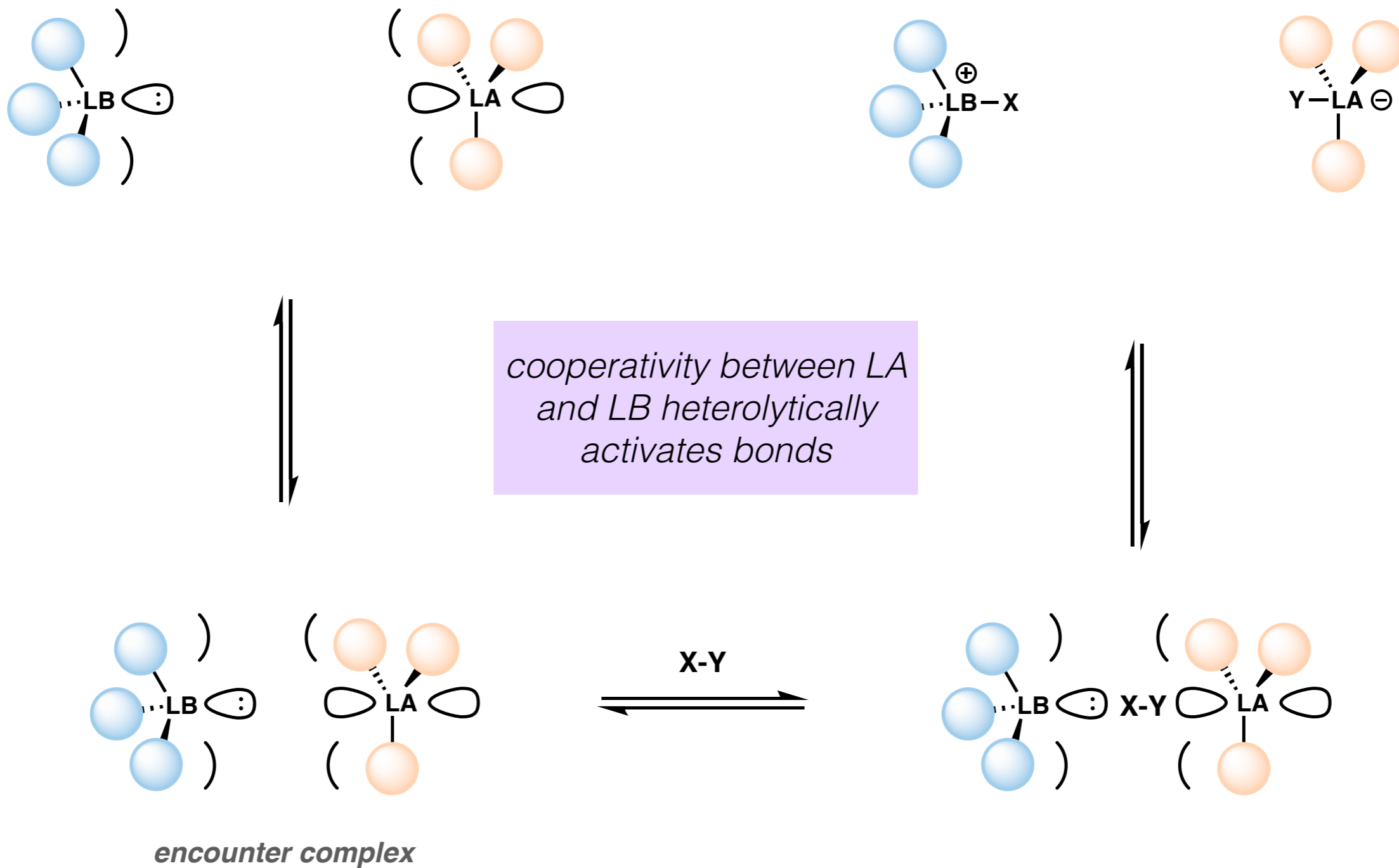
Electric Field Model

H₂ entry into cavity of FLP

Mechanism and General Reactivity of FLPs



Mechanism and General Reactivity of FLPs



History: Early Developments

Lewis, 1923



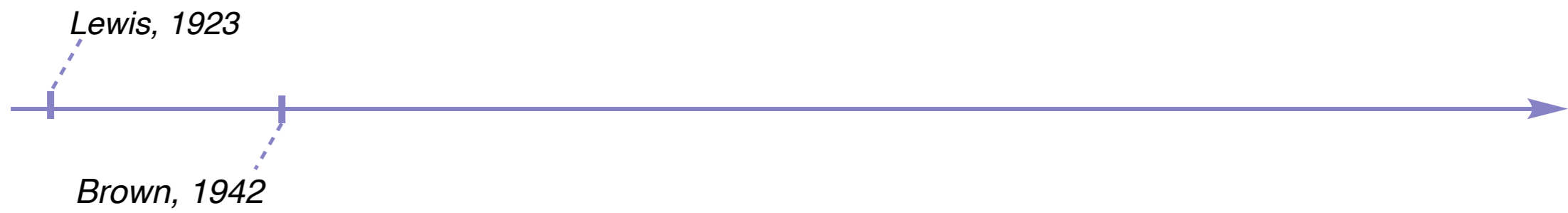
History: Early Developments

Lewis, 1923

“It seems to me that with complete generality we may say that a basic substance is one which has a lone pair of electrons which may be used to complete the stable group of another atom, and that an acid substance is one which can employ a lone pair from another molecule in completing the stable group of one of its own atoms. In other words, the basic substance furnishes a pair of electrons for a chemical bond, the acid substance accepts such a pair.”

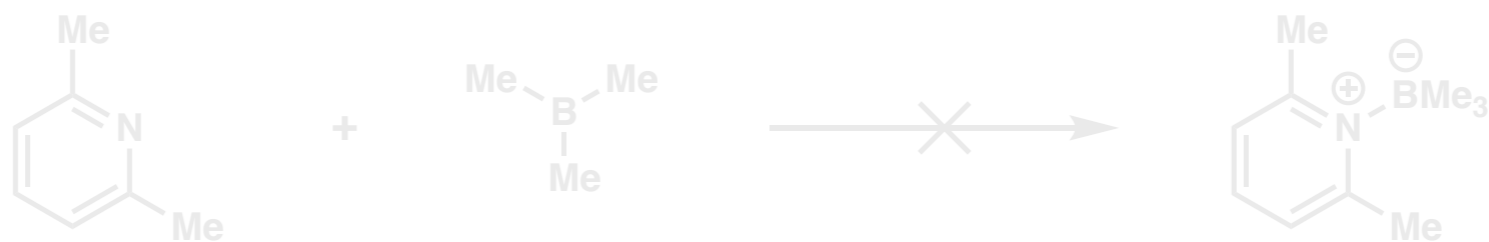
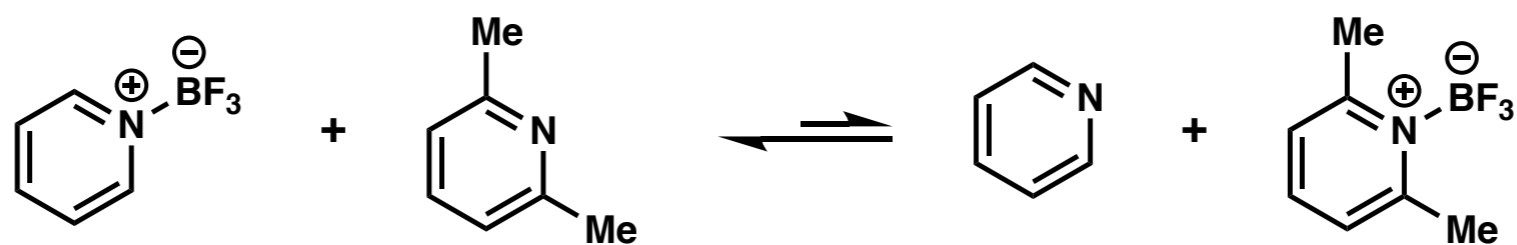


History: Early Developments



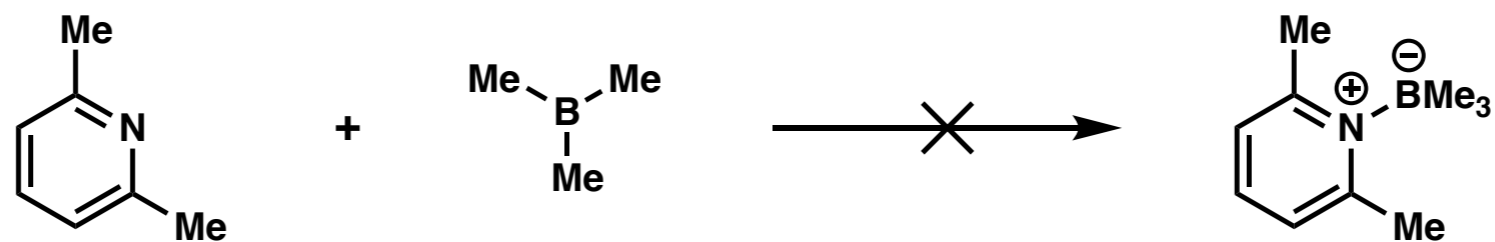
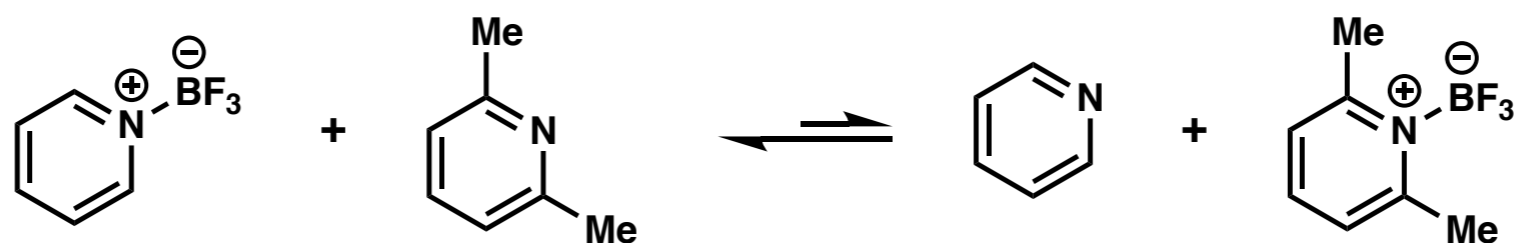
History: Early Developments

Brown, 1942



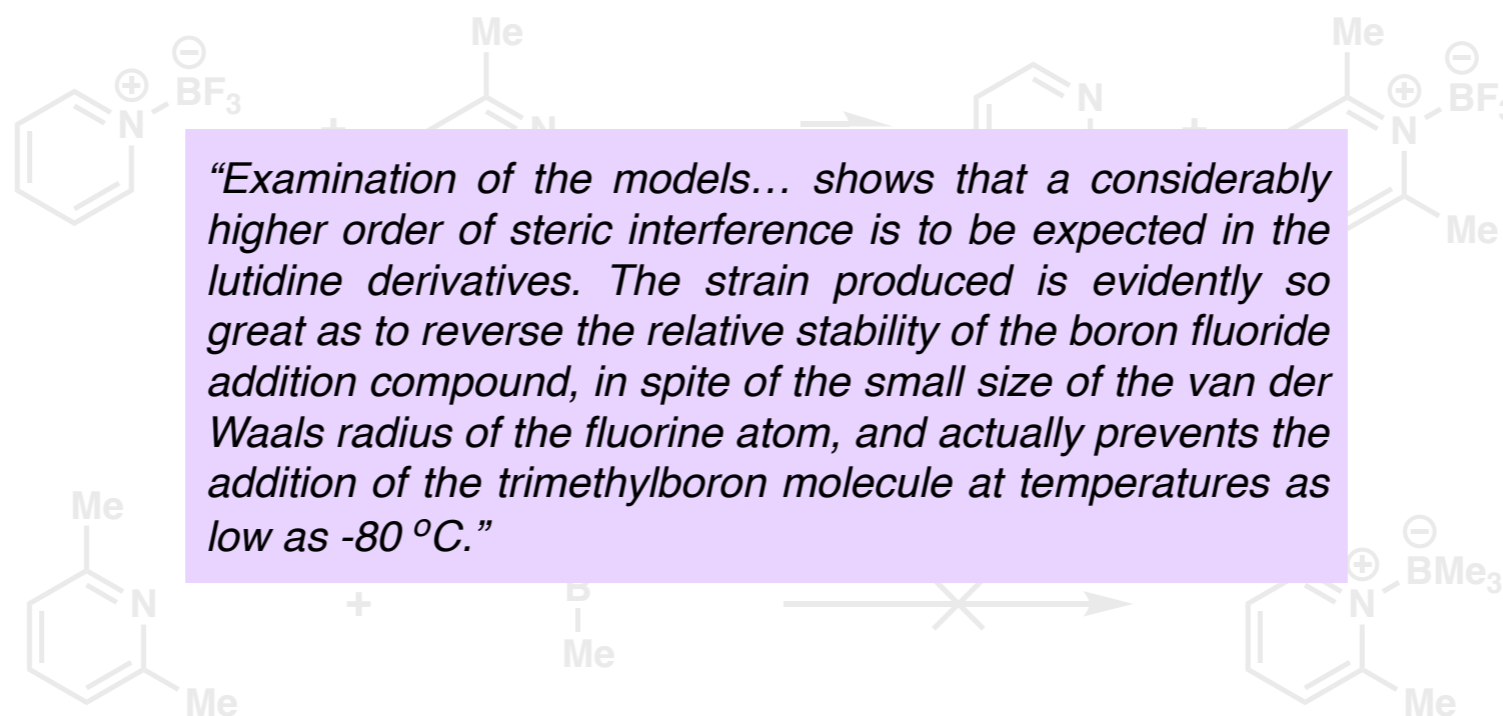
History: Early Developments

Brown, 1942

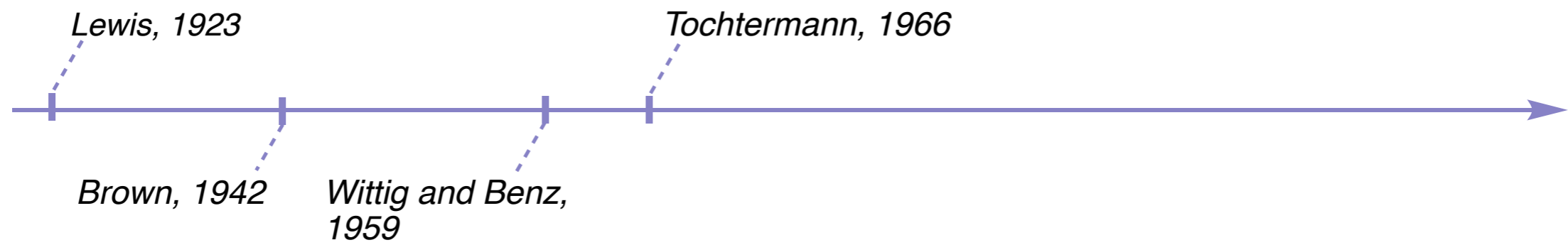


History: Early Developments

Brown, 1942

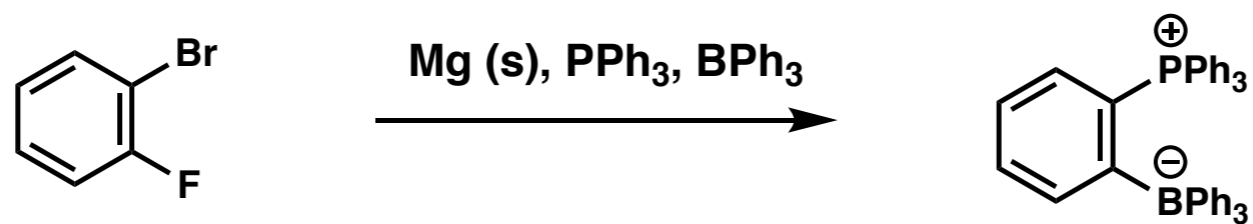


History: Early Developments



History: Early Developments

Wittig and Benz, 1959

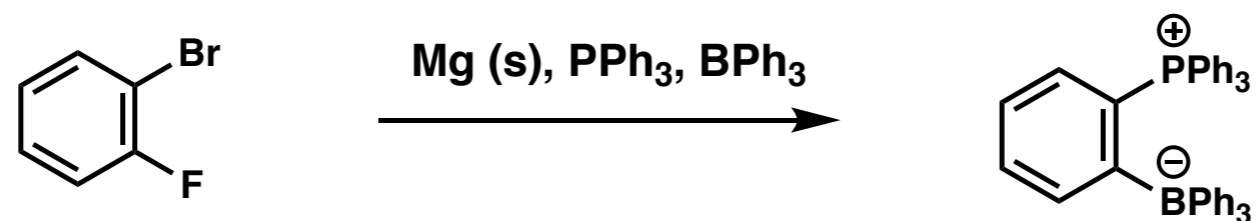


Tochtermann, 1966

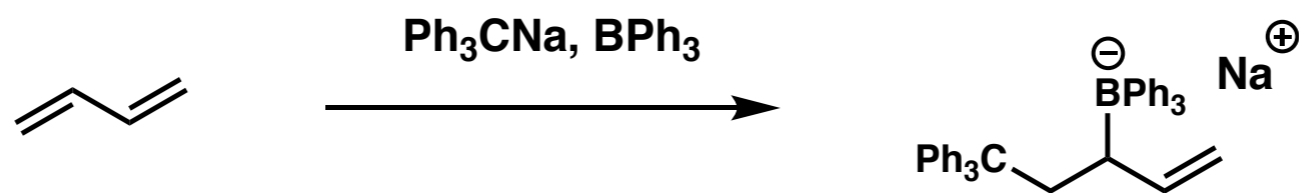


History: Early Developments

Wittig and Benz, 1959

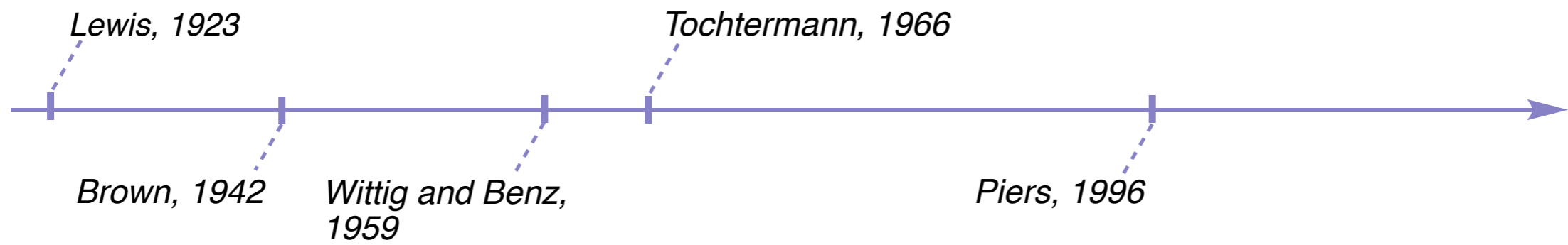


Tochtermann, 1966



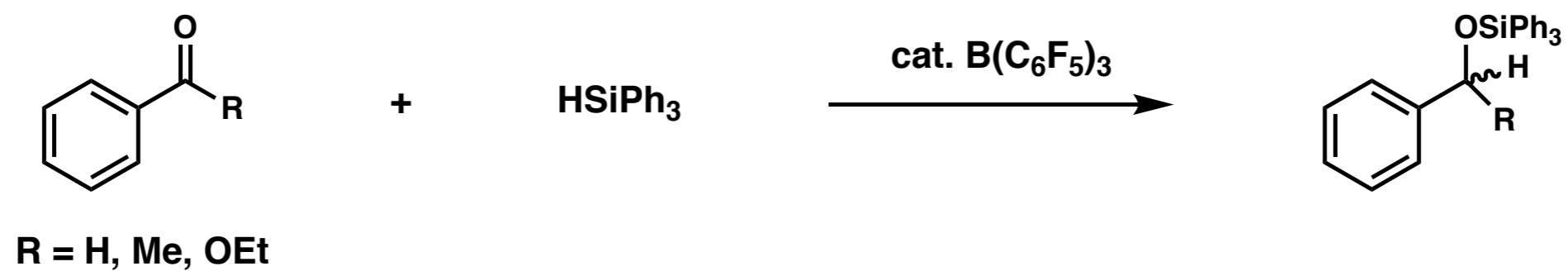
“Antagonistic Pairs”

History: Early Developments

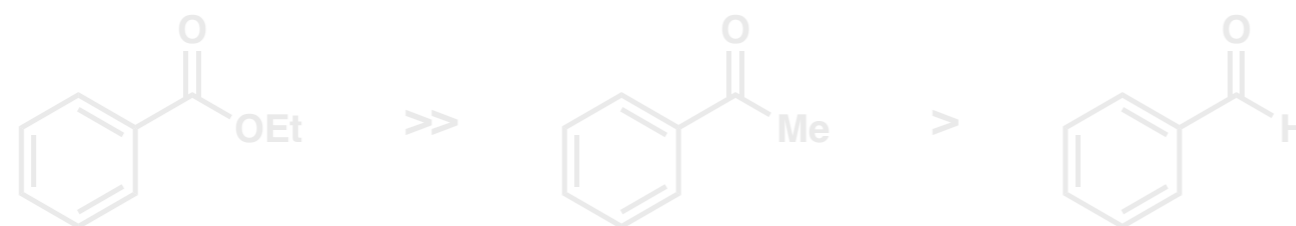


History: Early Developments

Piers, 1996

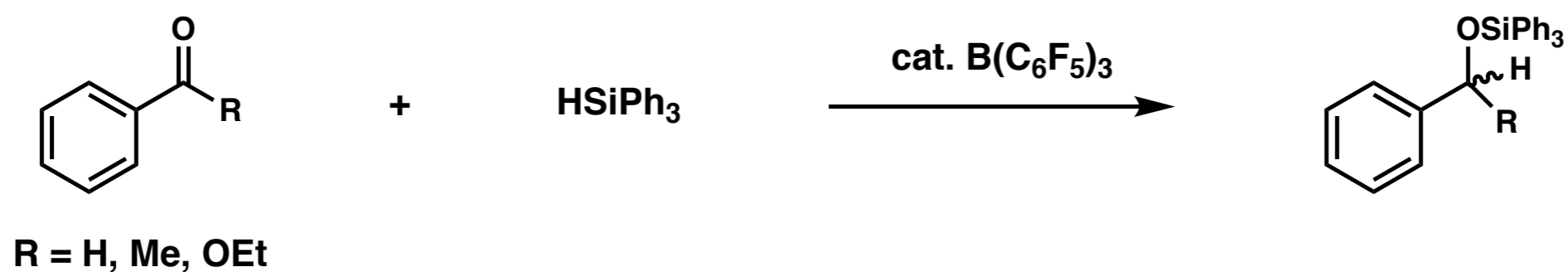


Observed Relative Rates

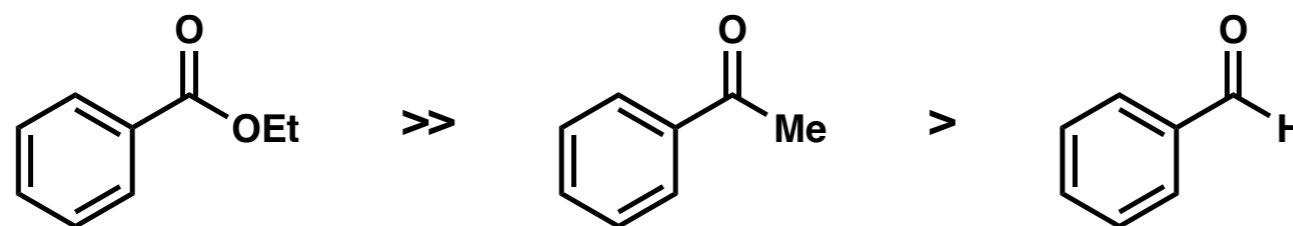


History: Early Developments

Piers, 1996

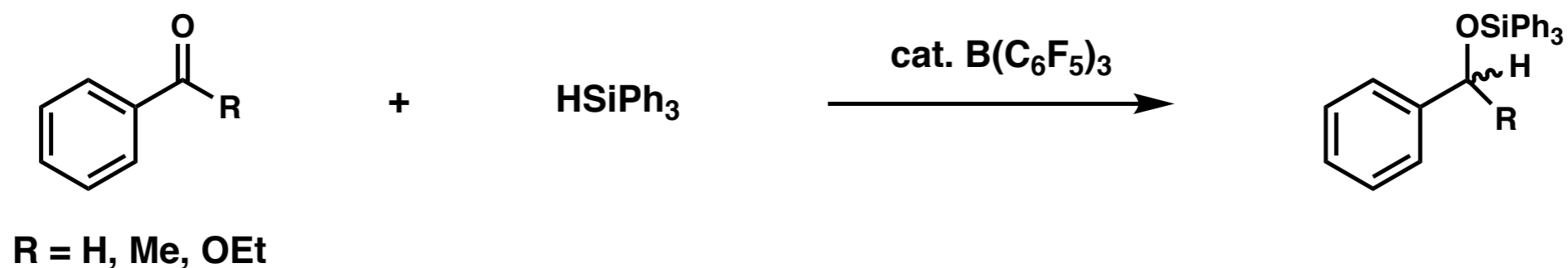


Observed Relative Rates

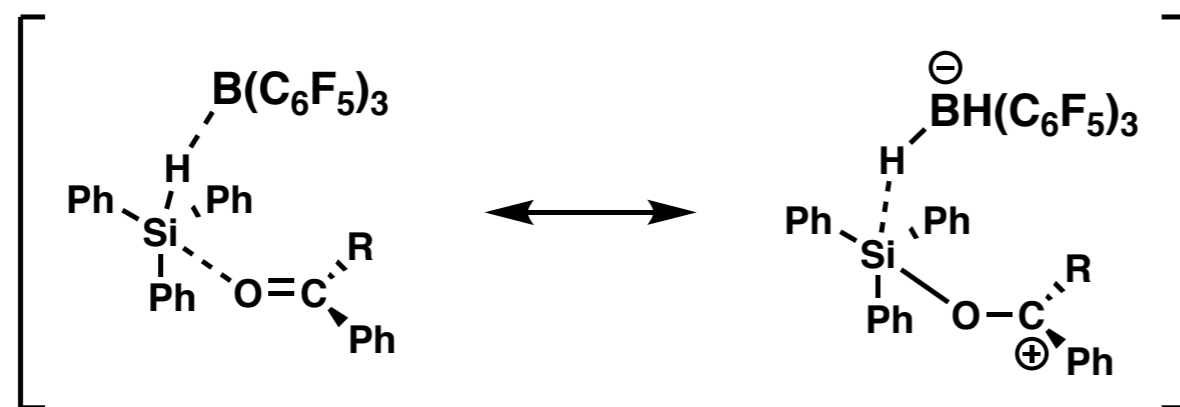


History: Early Developments

Piers, 1996

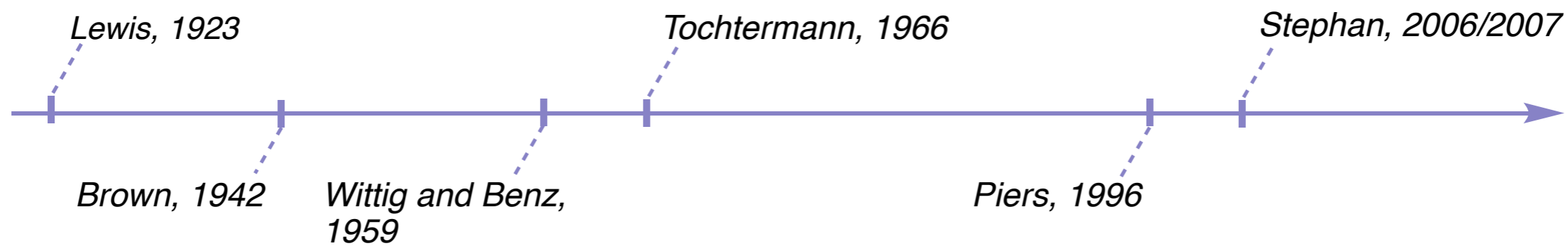


Proposed Mechanism



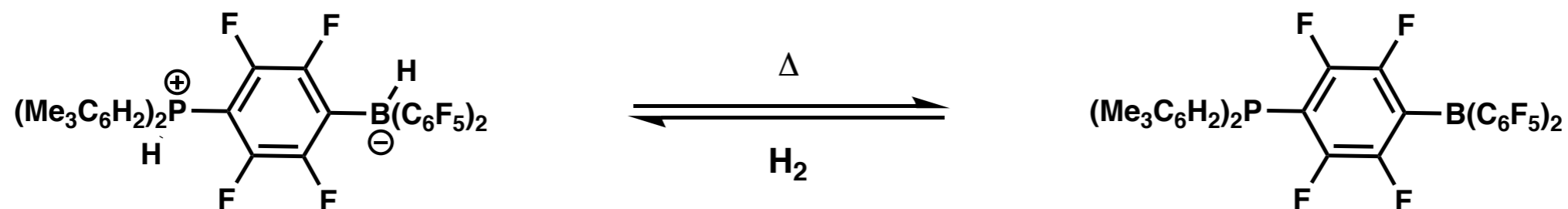
Si-H bond cleavage through activation by both Lewis acid (borane) and Lewis base (carbonyl)!

History: Seminal Publications

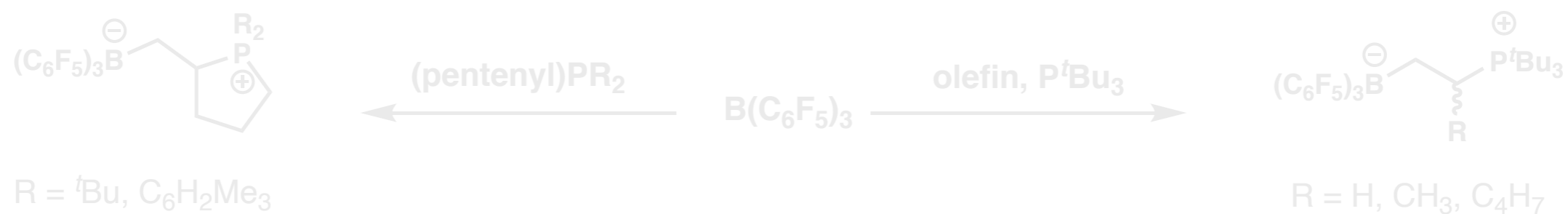


History: Seminal Publications

Stephan, 2006

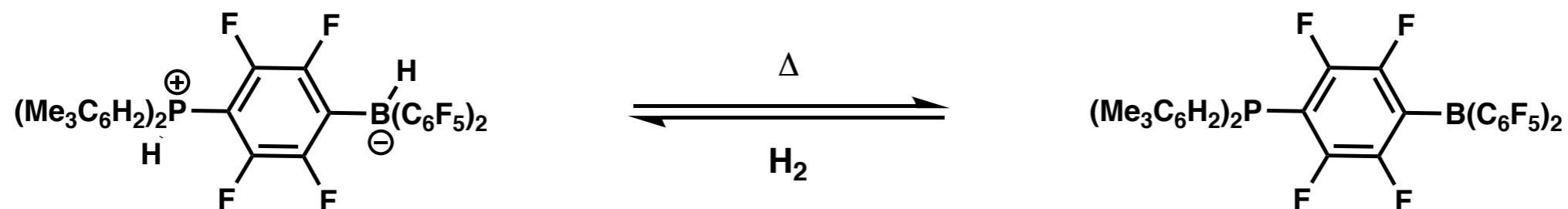


Stephan, 2007

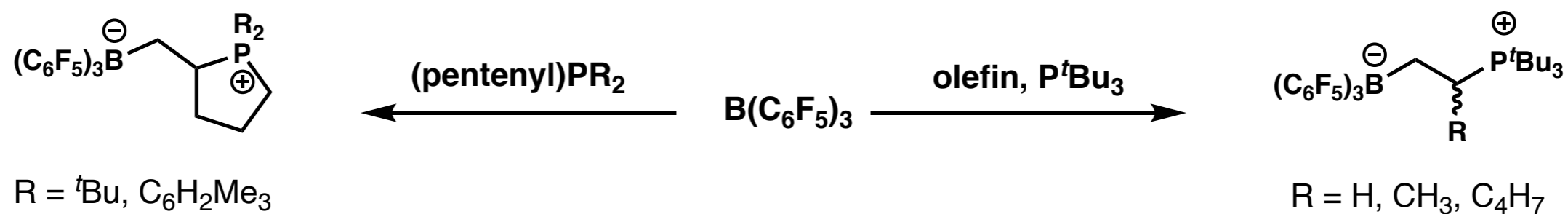


History: Seminal Publications

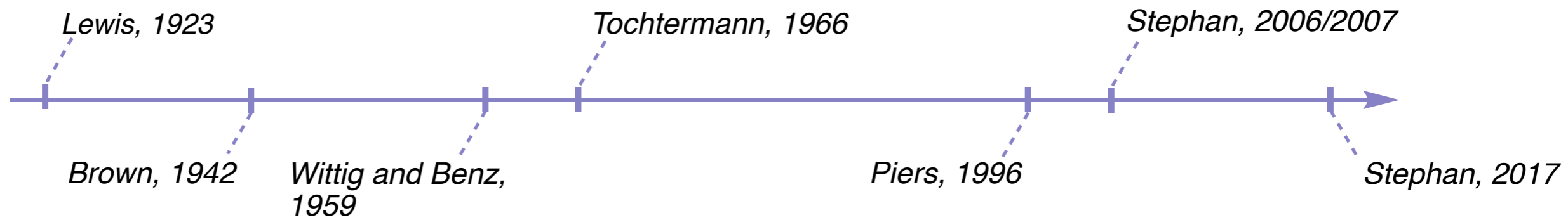
Stephan, 2006



Stephan, 2007

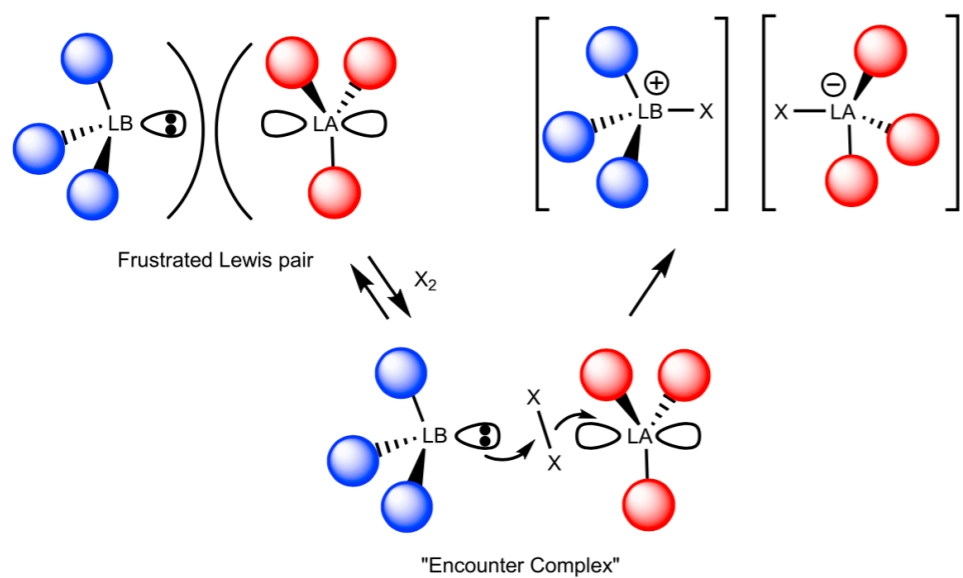


History: Seminal Publications



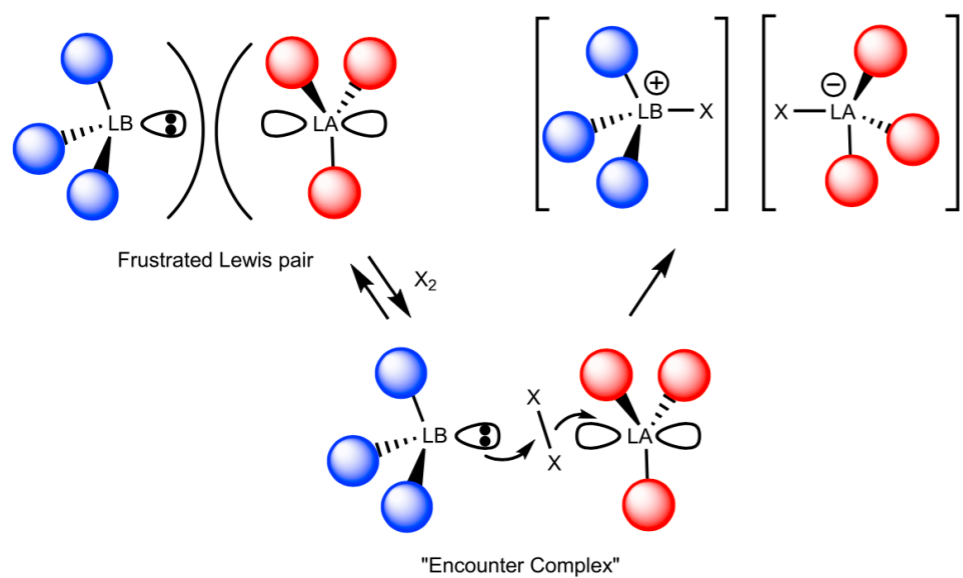
History: Seminal Publications

A Heterolytic Cleavage of X-X Bond

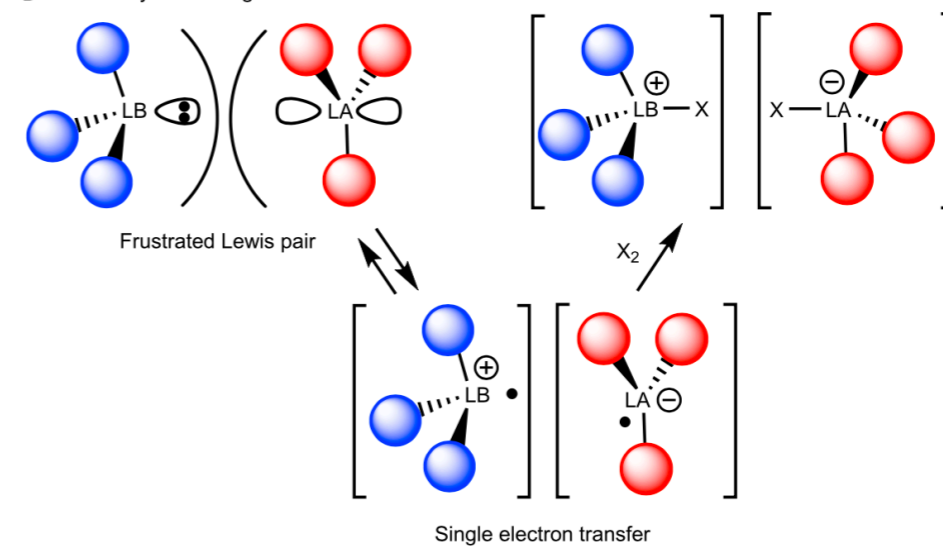


History: Seminal Publications

A Heterolytic Cleavage of X-X Bond

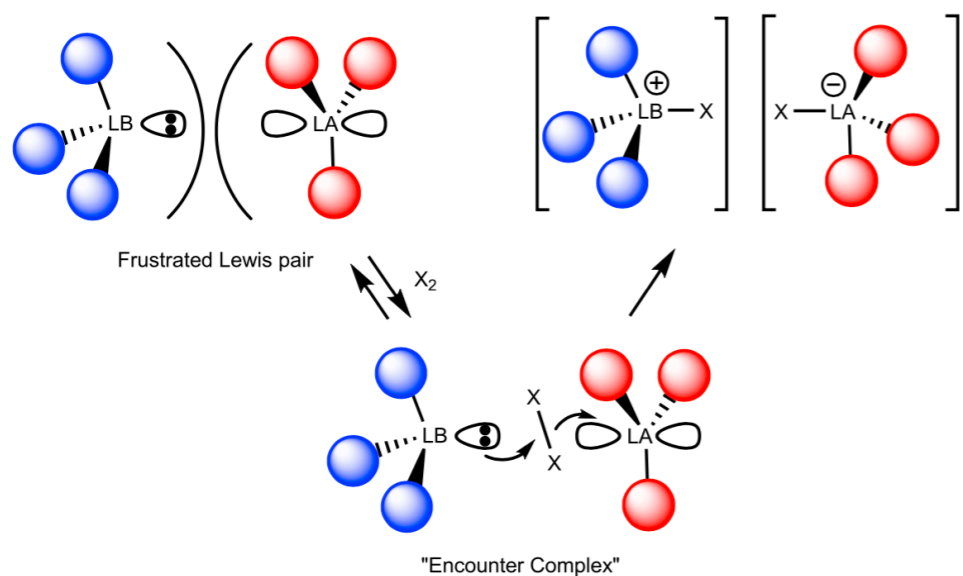


B Homolytic Cleavage of X-X Bond

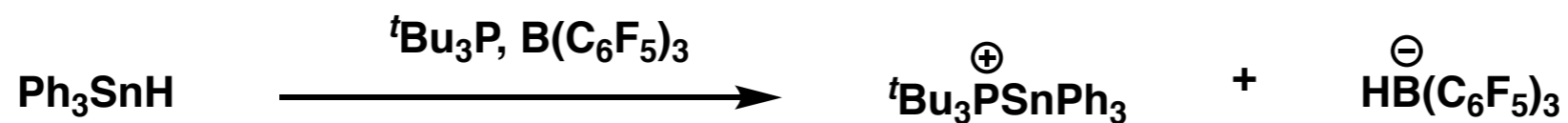
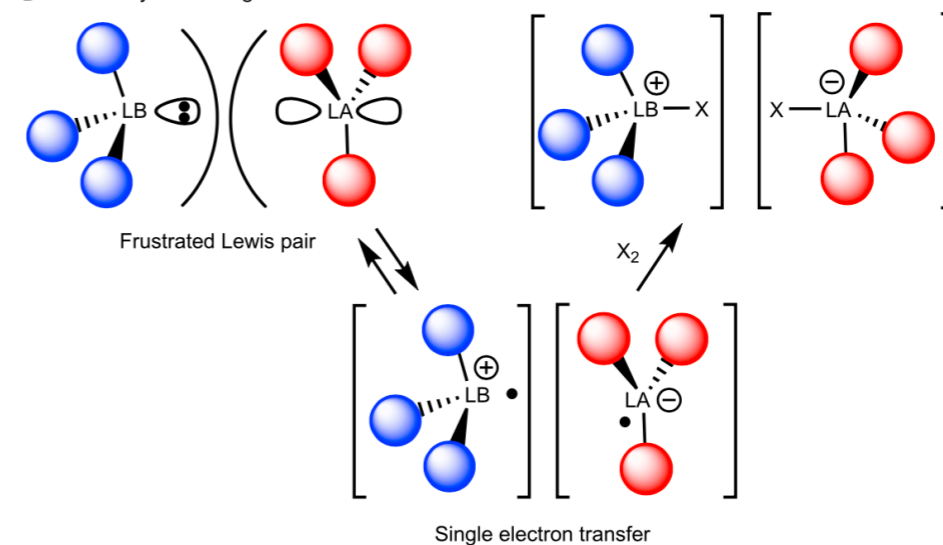


History: Seminal Publications

A Heterolytic Cleavage of X-X Bond

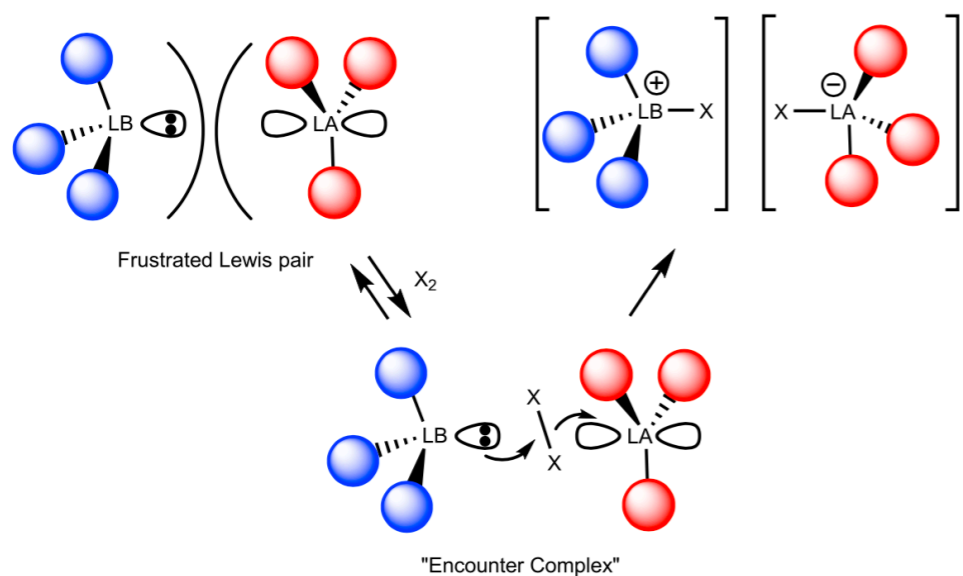


B Homolytic Cleavage of X-X Bond

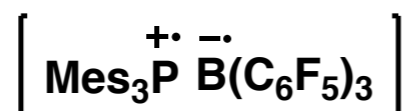
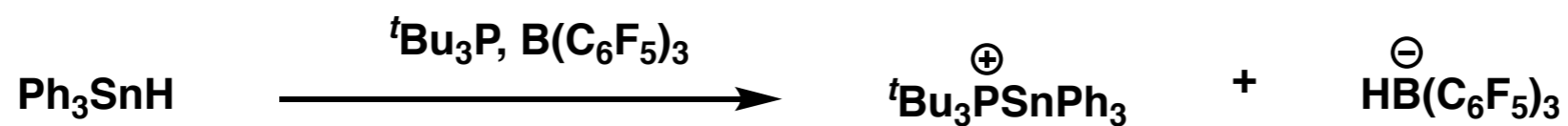
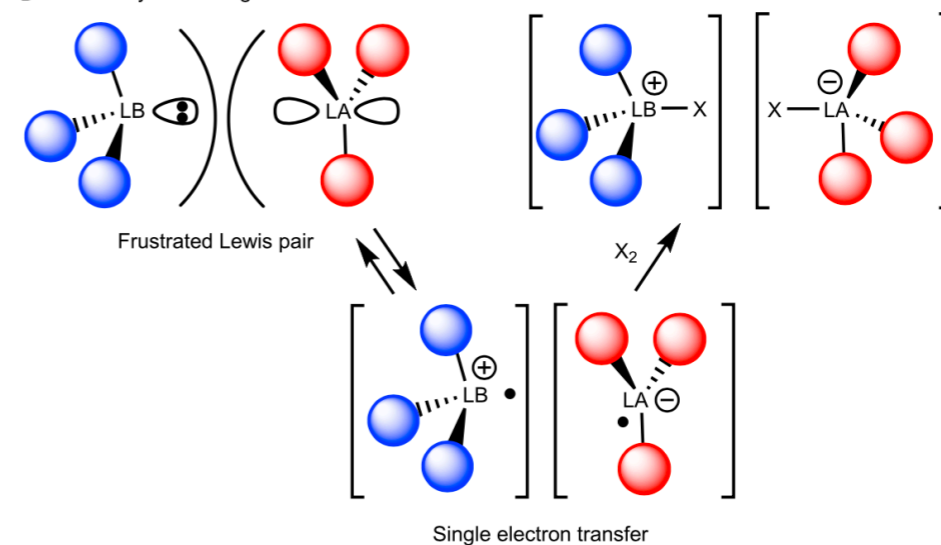


History: Seminal Publications

A Heterolytic Cleavage of X-X Bond



B Homolytic Cleavage of X-X Bond



Expanding Scope of FLPs

1 IA 1A												18 VIIIA 8A					
1 H Hydrogen 1.008	2 He Helium 4.003											13 IIIA 3A	14 IVA 4A	15 VA 5A	16 VIA 6A	17 VIIA 7A	
3 Li Lithium 6.941	4 Be Beryllium 9.012											5 B Boron 10.811	6 C Carbon 12.011	7 N Nitrogen 14.007	8 O Oxygen 15.999	9 F Fluorine 18.998	10 Ne Neon 20.180
11 Na Sodium 22.990	12 Mg Magnesium 24.305	3 IIIB 3B	4 IVB 4B	5 VB 5B	6 VIB 6B	7 VIIB 7B	8 VIII 8	9 VIII 8	10 VIII 8	11 IB 1B	12 IIB 2B	13 Al Aluminum 26.982	14 Si Silicon 28.086	15 P Phosphorus 30.974	16 S Sulfur 32.066	17 Cl Chlorine 35.453	18 Ar Argon 39.948
19 K Potassium 39.098	20 Ca Calcium 40.078	21 Sc Scandium 44.956	22 Ti Titanium 47.867	23 V Vanadium 50.942	24 Cr Chromium 51.996	25 Mn Manganese 54.938	26 Fe Iron 55.845	27 Co Cobalt 58.933	28 Ni Nickel 58.693	29 Cu Copper 63.546	30 Zn Zinc 65.38	31 Ga Gallium 69.723	32 Ge Germanium 72.631	33 As Arsenic 74.922	34 Se Selenium 78.971	35 Br Bromine 79.904	36 Kr Krypton 84.798
37 Rb Rubidium 84.468	38 Sr Strontium 87.62	39 Y Yttrium 88.906	40 Zr Zirconium 91.224	41 Nb Niobium 92.906	42 Mo Molybdenum 95.95	43 Tc Technetium 98.907	44 Ru Ruthenium 101.07	45 Rh Rhodium 102.906	46 Pd Palladium 106.42	47 Ag Silver 107.868	48 Cd Cadmium 112.414	49 In Indium 114.818	50 Sn Tin 118.711	51 Sb Antimony 121.760	52 Te Tellurium 127.6	53 I Iodine 126.904	54 Xe Xenon 131.294
55 Cs Cesium 132.905	56 Ba Barium 137.328	57-71	72 Hf Hafnium 178.49	73 Ta Tantalum 180.948	74 W Tungsten 183.84	75 Re Rhenium 186.207	76 Os Osmium 190.23	77 Ir Iridium 192.217	78 Pt Platinum 195.085	79 Au Gold 196.967	80 Hg Mercury 200.592	81 Tl Thallium 204.383	82 Pb Lead 207.2	83 Bi Bismuth 208.980	84 Po Polonium [208.982]	85 At Astatine 209.987	86 Rn Radon 222.018
87 Fr Francium 223.020	88 Ra Radium 226.025	89-103	104 Rf Rutherfordium [261]	105 Db Dubnium [262]	106 Sg Seaborgium [266]	107 Bh Bohrium [264]	108 Hs Hassium [269]	109 Mt Meitnerium [268]	110 Ds Darmstadtium [269]	111 Rg Roentgenium [272]	112 Cn Copernicium [277]	113 Nh Nihonium unknown	114 Fl Flerovium [289]	115 Mc Moscovium unknown	116 Lv Livermorium [298]	117 Ts Tennessine unknown	118 Og Oganesson unknown

Lanthanide Series	57 La Lanthanum 138.905	58 Ce Cerium 140.116	59 Pr Praseodymium 140.908	60 Nd Neodymium 144.243	61 Pm Promethium 144.913	62 Sm Samarium 150.36	63 Eu Europium 151.964	64 Gd Gadolinium 157.25	65 Tb Terbium 158.925	66 Dy Dysprosium 162.500	67 Ho Holmium 164.930	68 Er Erbium 167.259	69 Tm Thulium 168.934	70 Yb Ytterbium 173.055	71 Lu Lutetium 174.967
Actinide Series	89 Ac Actinium 227.028	90 Th Thorium 232.038	91 Pa Protactinium 231.036	92 U Uranium 238.029	93 Np Neptunium 237.048	94 Pu Plutonium 244.064	95 Am Americium 243.061	96 Cm Curium 247.070	97 Bk Berkelium 247.070	98 Cf Californium 251.080	99 Es Einsteinium [254]	100 Fm Fermium 257.095	101 Md Mendelevium 258.1	102 No Nobelium 259.101	103 Lr Lawrencium [262]

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Expanding Scope of FLPs

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37 Rb Rubidium 84.468	38 Sr Strontium 87.62	39 Y Yttrium 88.906	40 Zr Zirconium 91.224	41 Nb Niobium 92.906	42 Mo Molybdenum 95.95	43 Tc Technetium 98.907	44 Ru Ruthenium 101.07	45 Rh Rhodium 102.906	46 Pd Palladium 106.42	47 Ag Silver 107.868	48 Cd Cadmium 112.414	49 In Indium 114.818	50 Sn Tin 118.711	51 Sb Antimony 121.760	52 Te Tellurium 127.6	53 I Iodine 126.904	54 Xe Xenon 131.294
55 Cs Cesium 132.905	56 Ba Barium 137.328	57-71	72 Hf Hafnium 178.49	73 Ta Tantalum 180.948	74 W Tungsten 183.84	75 Re Rhenium 186.207	76 Os Osmium 190.23	77 Ir Iridium 192.217	78 Pt Platinum 195.085	79 Au Gold 196.967	80 Hg Mercury 200.592	81 Tl Thallium 204.383	82 Pb Lead 207.2	83 Bi Bismuth 208.980	84 Po Polonium [208.982]	85 At Astatine 209.987	86 Rn Radon 222.018
87 Fr Francium 223.020	88 Ra Radium 226.025	89-103	104 Rf Rutherfordium [261]	105 Db Dubnium [262]	106 Sg Seaborgium [266]	107 Bh Bohrium [264]	108 Hs Hassium [269]	109 Mt Meitnerium [268]	110 Ds Darmstadtium [269]	111 Rg Roentgenium [272]	112 Cn Copernicium [277]	113 Nh Nihonium unknown	114 Fl Flerovium [289]	115 Mc Moscovium unknown	116 Lv Livermorium [298]	117 Ts Tennessine unknown	118 Og Oganesson unknown

Lanthanide Series	57 La Lanthanum 138.905	58 Ce Cerium 140.116	59 Pr Praseodymium 140.908	60 Nd Neodymium 144.243	61 Pm Promethium 144.913	62 Sm Samarium 150.36	63 Eu Europium 151.964	64 Gd Gadolinium 157.25	65 Tb Terbium 158.925	66 Dy Dysprosium 162.500	67 Ho Holmium 164.930	68 Er Erbium 167.259	69 Tm Thulium 168.934	70 Yb Ytterbium 173.055	71 Lu Lutetium 174.967
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Expanding Scope of FLPs

1 IA 1A																	18 VIIIA 8A	
1 H Hydrogen 1.008																		2 He Helium 4.003
3 Li Lithium 6.941	4 Be Beryllium 9.012											5 B Boron 10.811	6 C Carbon 12.011	7 N Nitrogen 14.007	8 O Oxygen 15.999	9 F Fluorine 18.998	10 Ne Neon 20.180	
11 Na Sodium 22.990	12 Mg Magnesium 24.305	3 IIIB 3B	4 IVB 4B	5 VB 5B	6 VIB 6B	7 VIIB 7B	8 VIII 8	9 VIII 8	10 VIII 8	11 IB 1B	12 IIB 2B	13 Al Aluminum 26.982	14 Si Silicon 28.086	15 P Phosphorus 30.974	16 S Sulfur 32.066	17 Cl Chlorine 35.453	18 Ar Argon 39.948	
19 K Potassium 39.098	20 Ca Calcium 40.078	21 Sc Scandium 44.956	22 Ti Titanium 47.867	23 V Vanadium 50.942	24 Cr Chromium 51.996	25 Mn Manganese 54.938	26 Fe Iron 55.845	27 Co Cobalt 58.933	28 Ni Nickel 58.693	29 Cu Copper 63.546	30 Zn Zinc 65.38	31 Ga Gallium 69.723	32 Ge Germanium 72.631	33 As Arsenic 74.922	34 Se Selenium 78.971	35 Br Bromine 79.904	36 Kr Krypton 84.798	
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Expanding Scope of FLPs

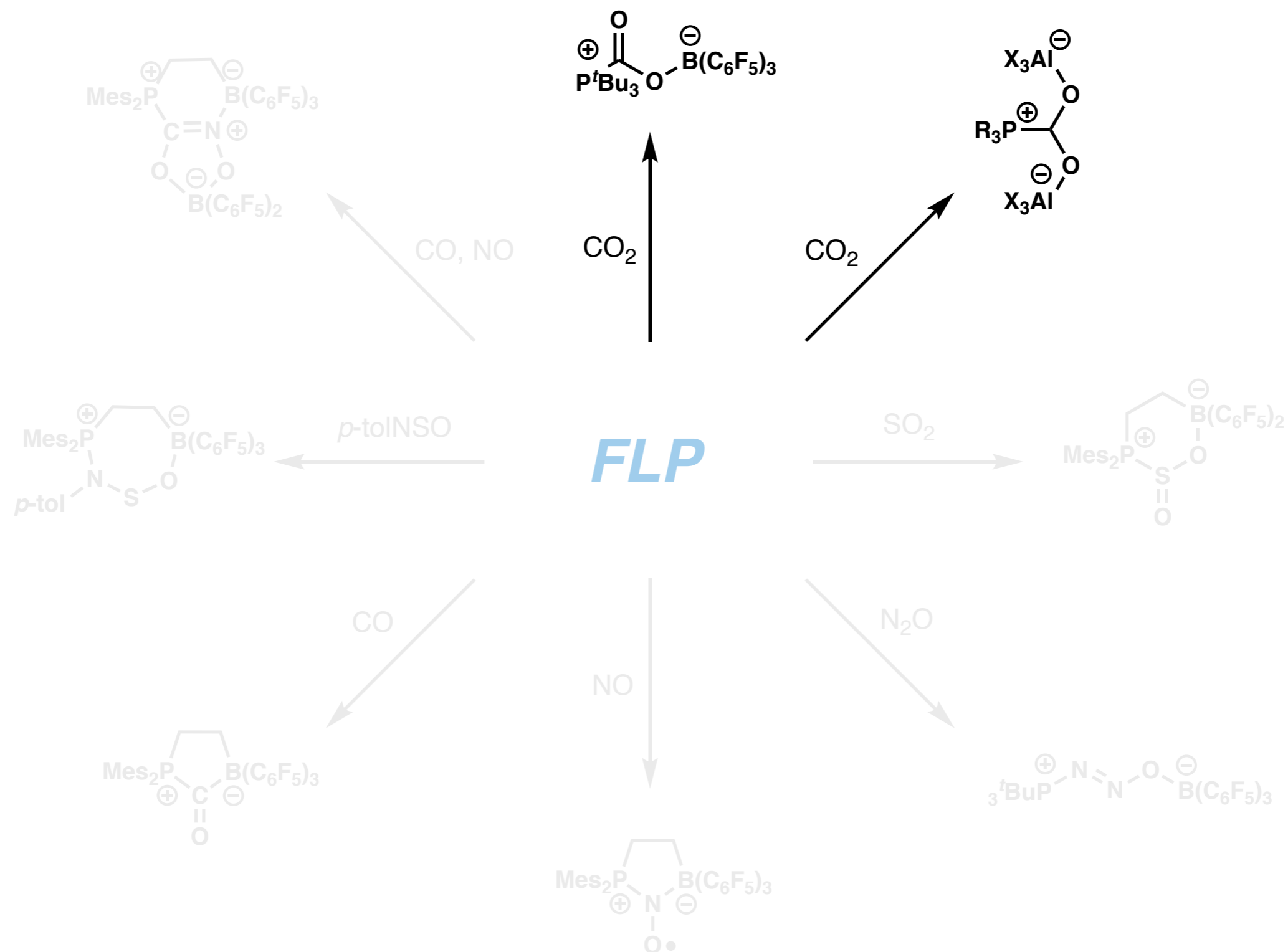
1 IA 1A																	18 VIIIA 8A
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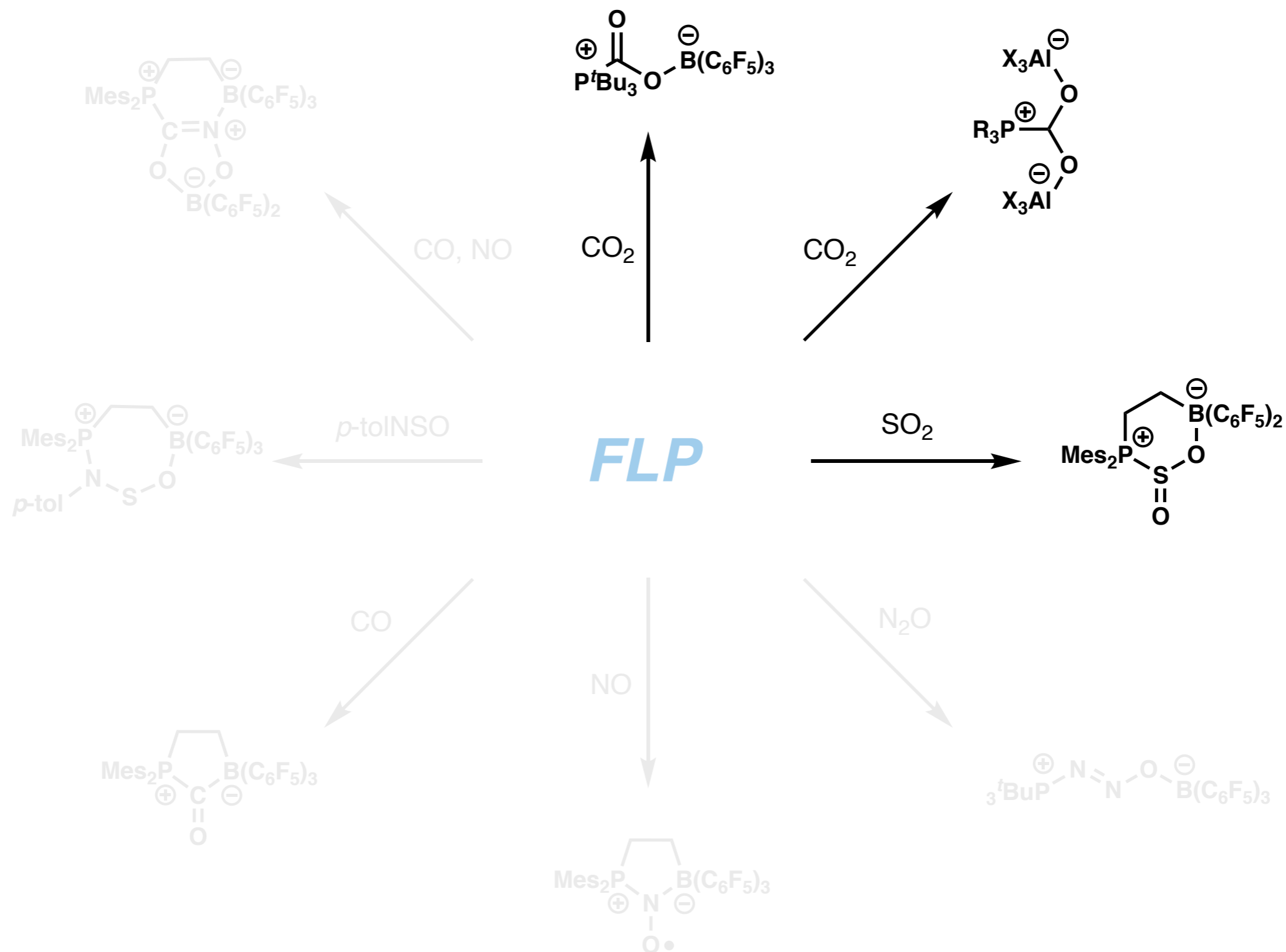
+ transition metals!



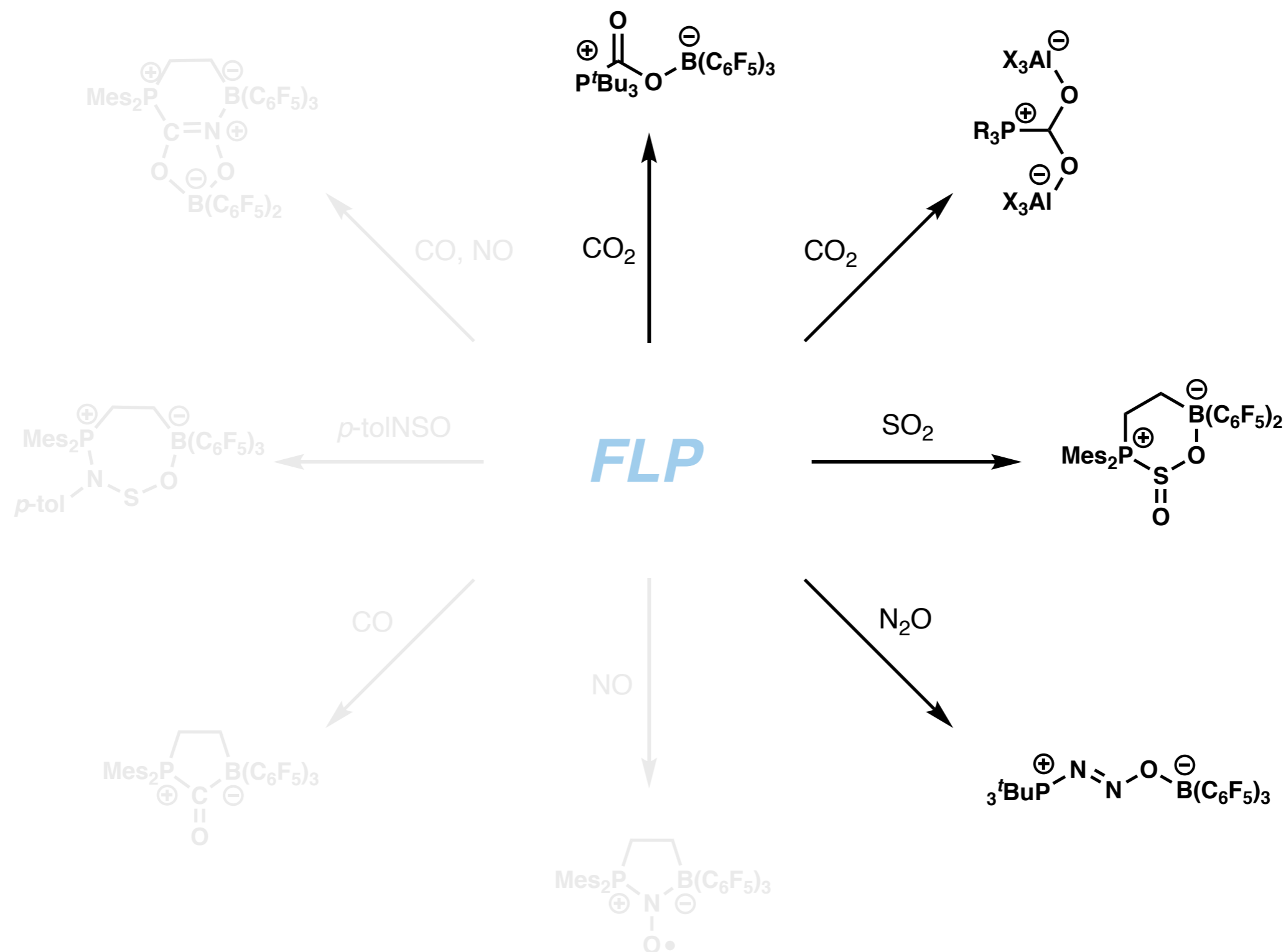
FLP Small Molecule Activation



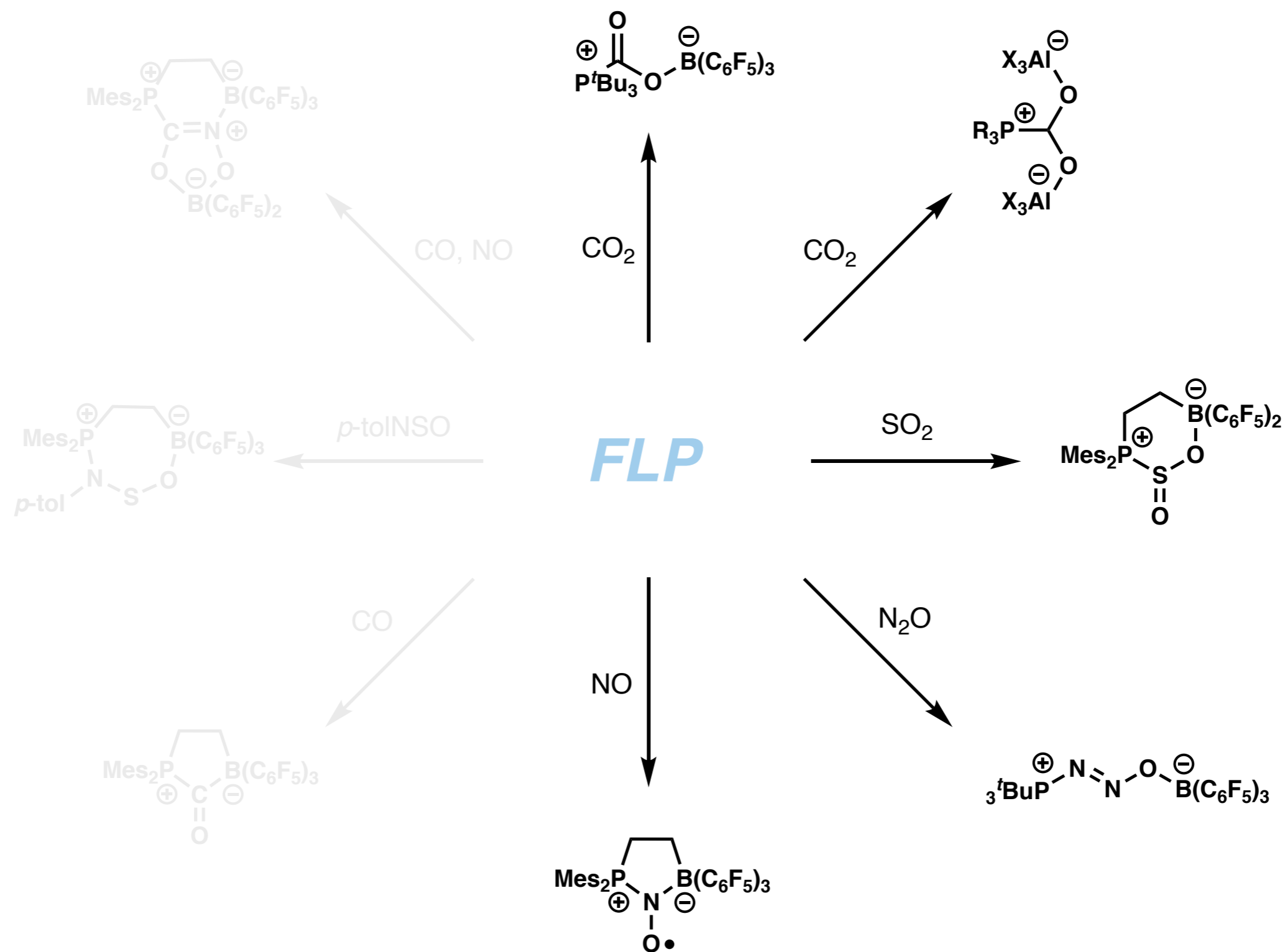
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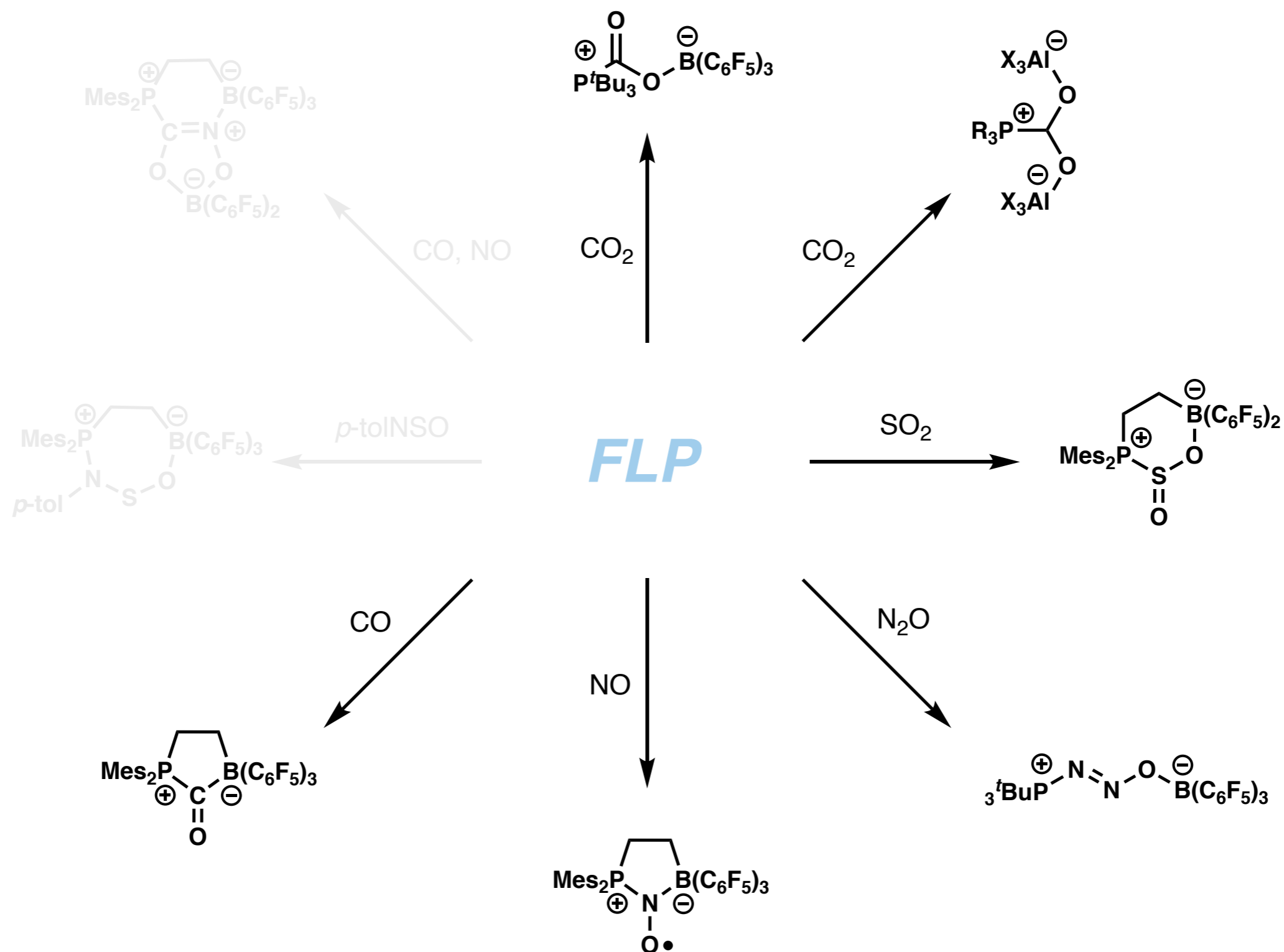
FLP Small Molecule Activation



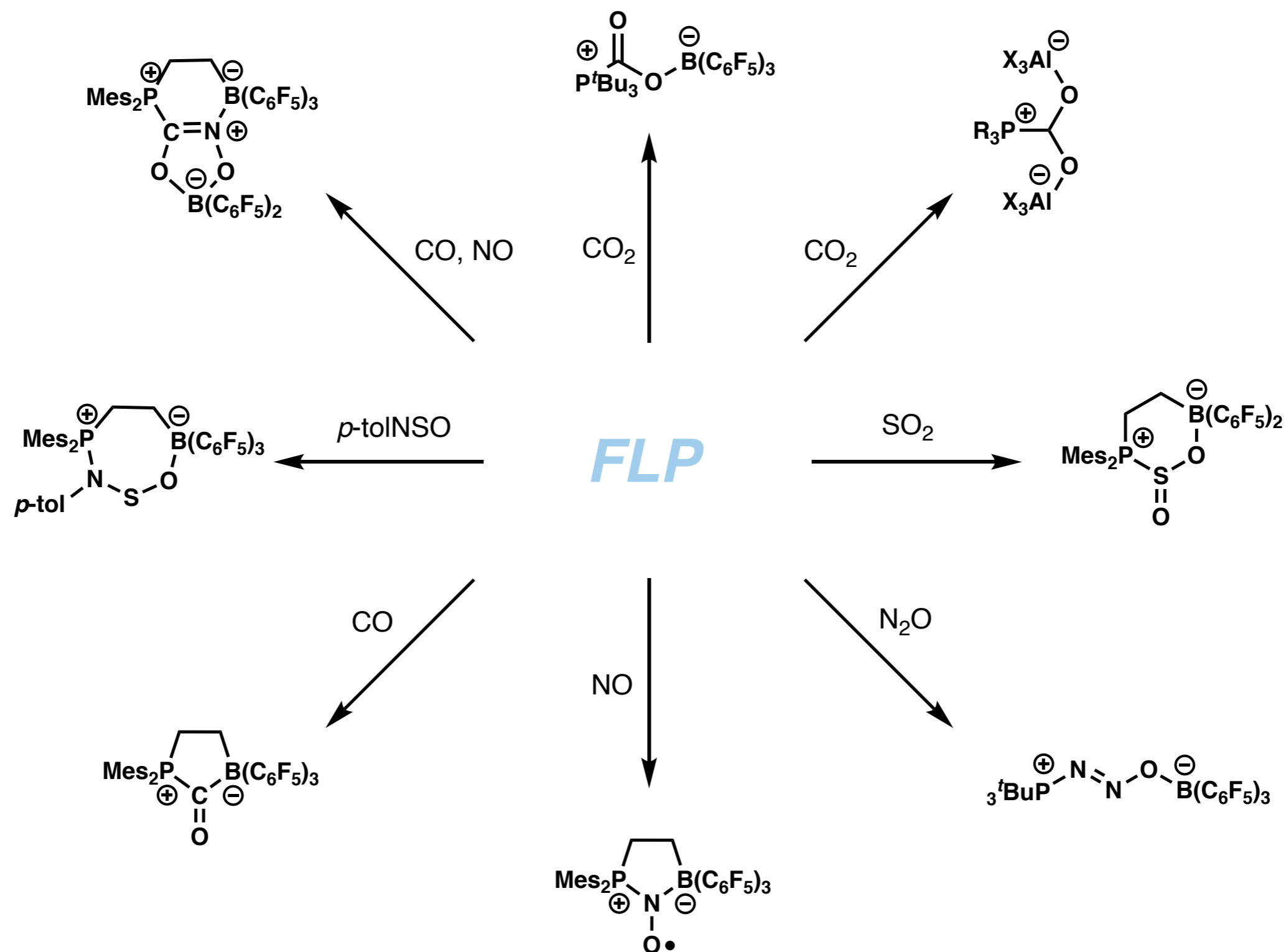
FLP Small Molecule Activation



FLP Small Molecule Activation



FLP Small Molecule Activation



Science, 2016, 354, 1248.

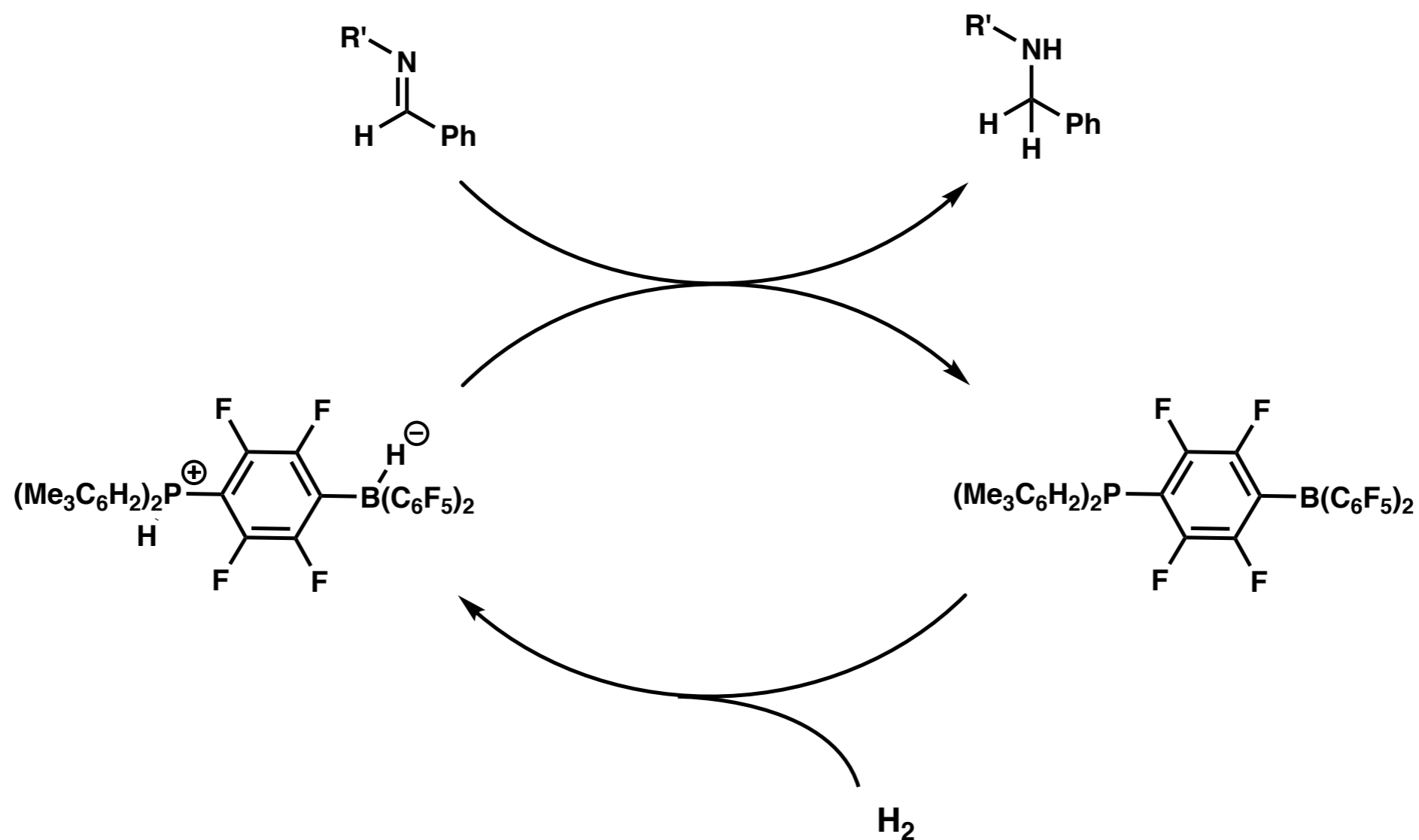
Angew. Chem. Int. Ed. 2015, 54, 809-812.

Angew. Chem. Int. Ed. 2016, 55, 9216-9219.

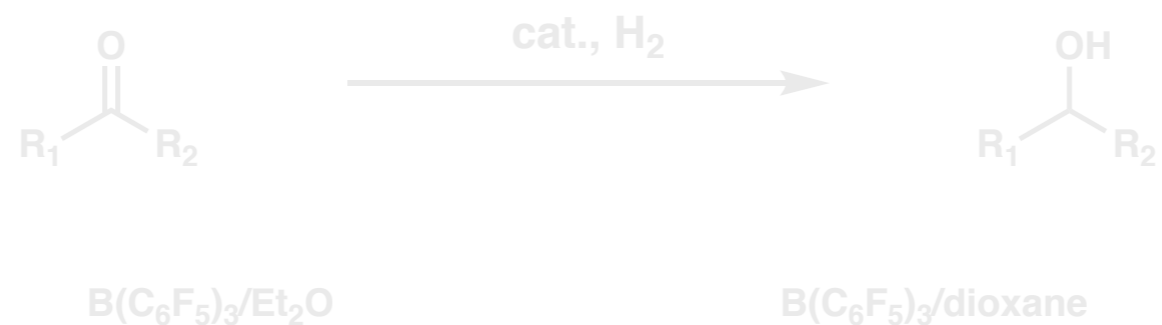
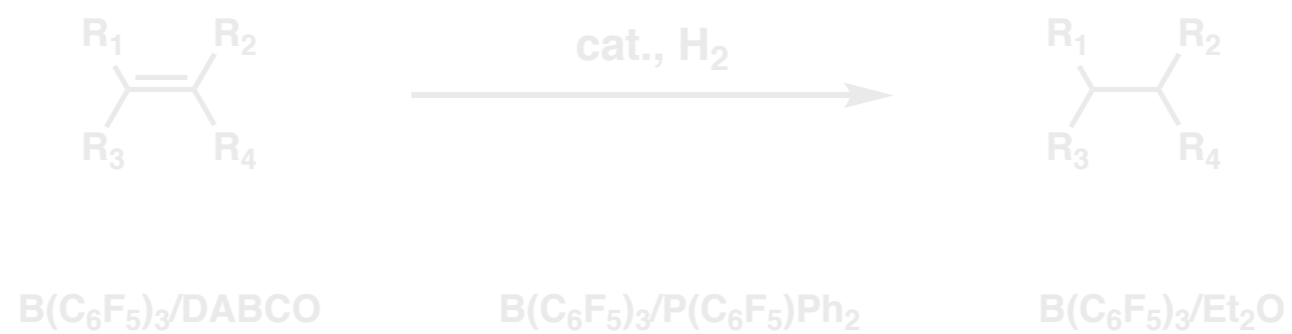
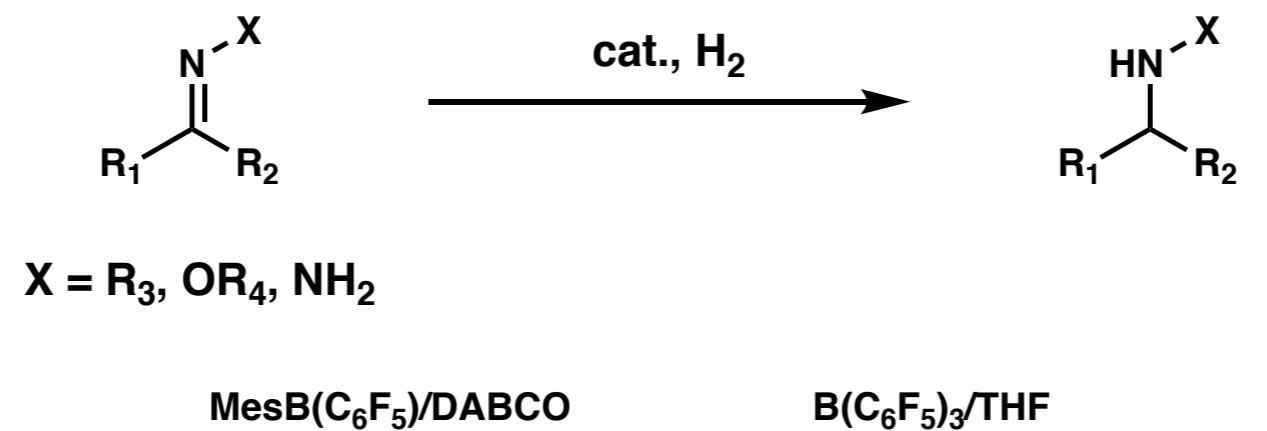


FLPs in Organic Transformations: Reductions

Stephan, 2007



FLPs in Organic Transformations: Reductions



B(C₆F₅)₃/DABCO

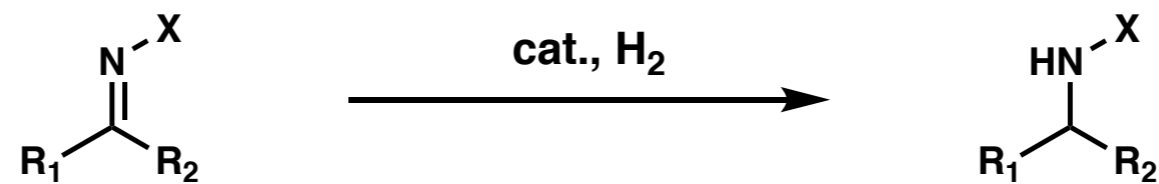
B(C₆F₅)₃/P(C₆F₅)Ph₂

B(C₆F₅)₃/Et₂O

B(C₆F₅)₃/Et₂O

B(C₆F₅)₃/dioxane

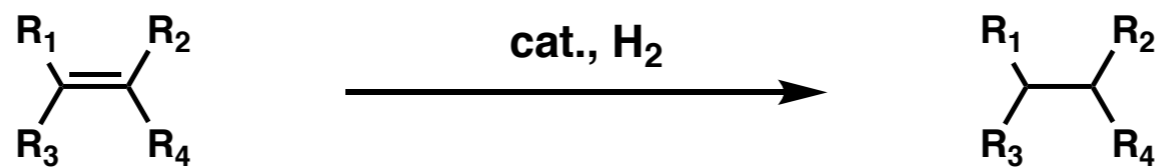
FLPs in Organic Transformations: Reductions



X = R₃, OR₄, NH₂

MesB(C₆F₅)/DABCO

B(C₆F₅)₃/THF



B(C₆F₅)₃/DABCO

B(C₆F₅)₃/P(C₆F₅)Ph₂

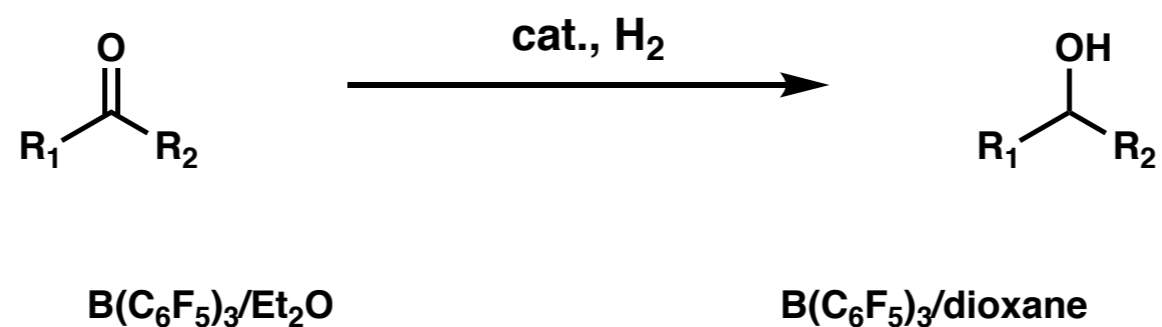
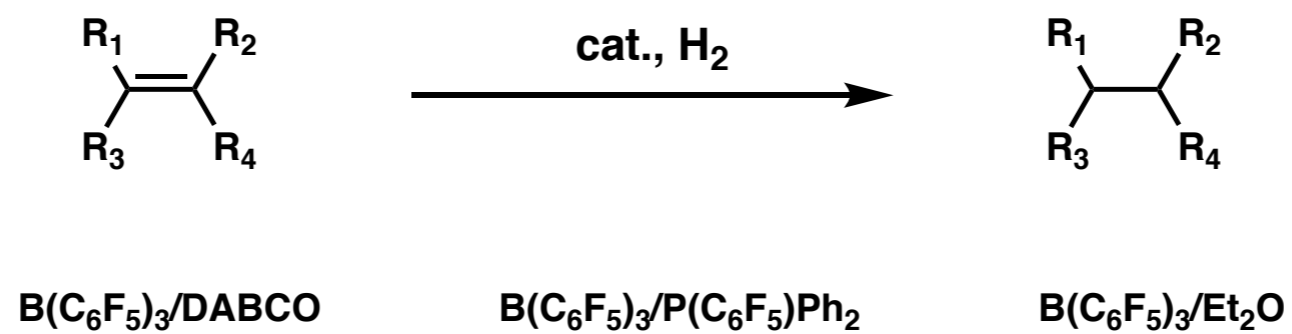
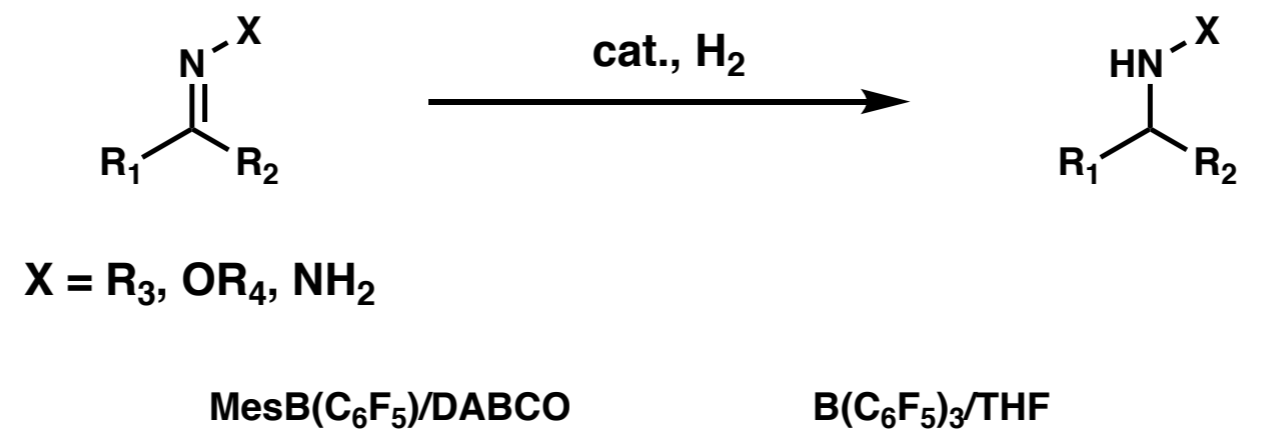
B(C₆F₅)₃/Et₂O



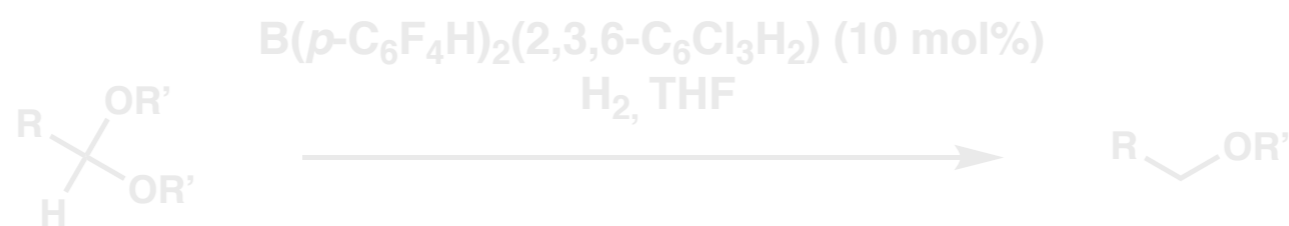
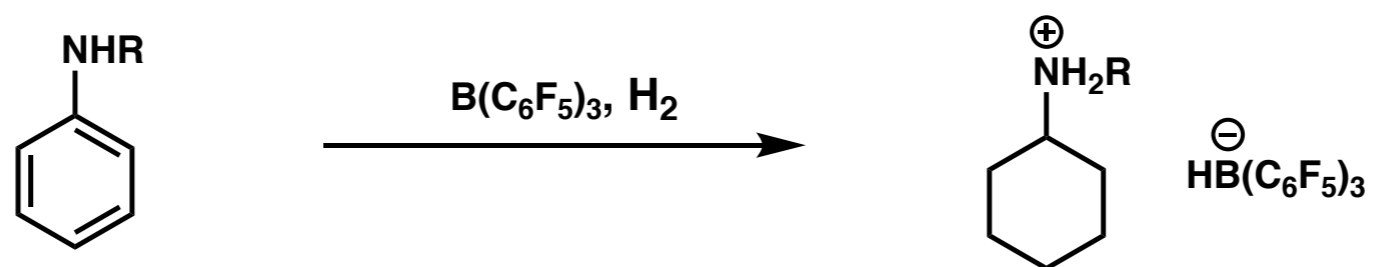
B(C₆F₅)₃/Et₂O

B(C₆F₅)₃/dioxane

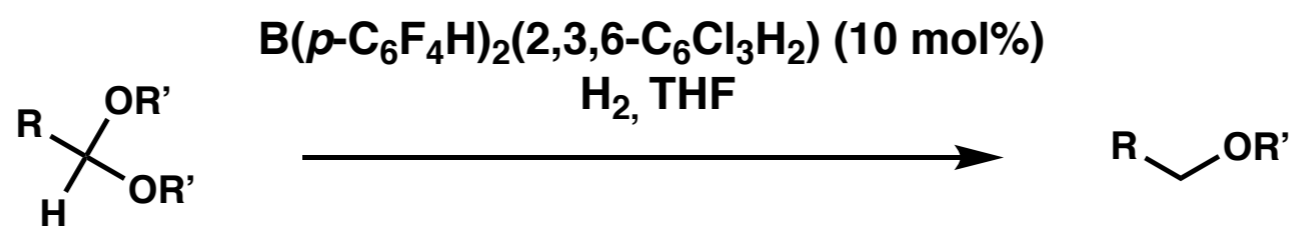
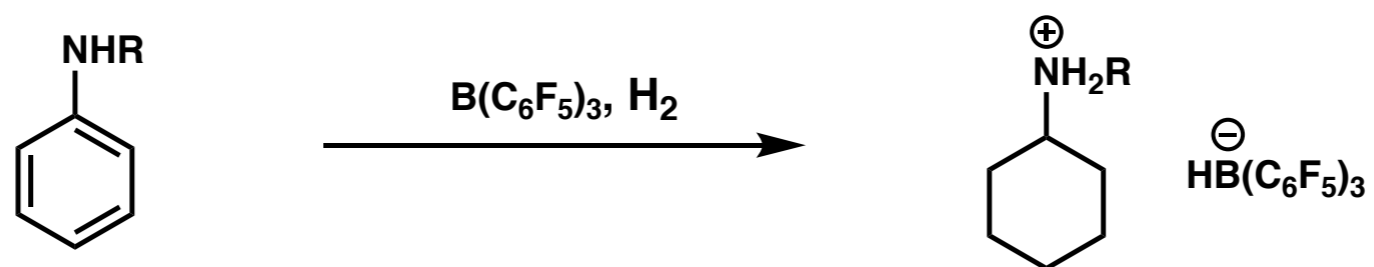
FLPs in Organic Transformations: Reductions



FLPs in Organic Transformations: Reductions

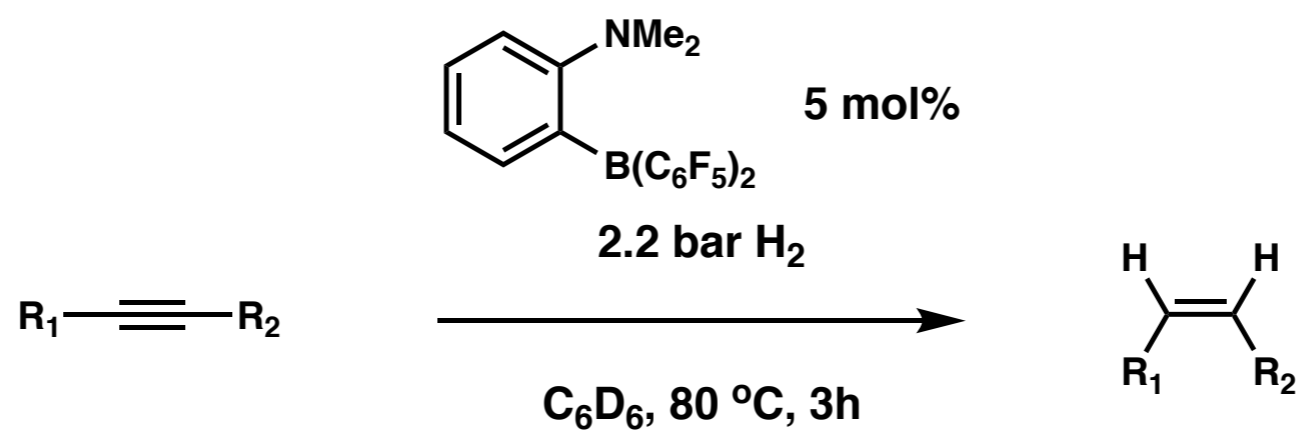


FLPs in Organic Transformations: Reductions



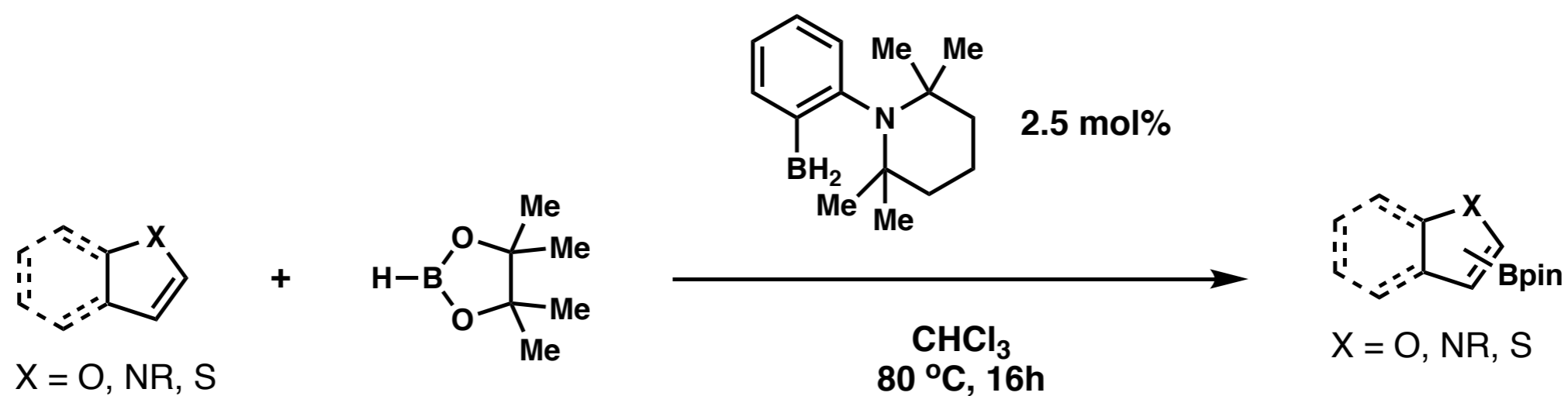
FLPs in Organic Transformations: Reductions

Repo, 2013



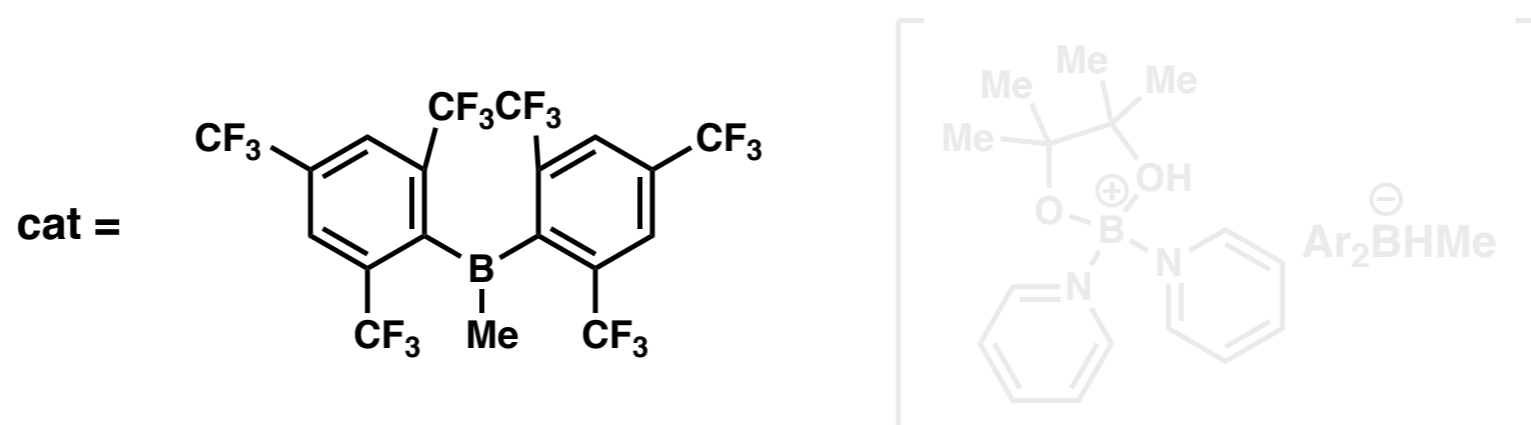
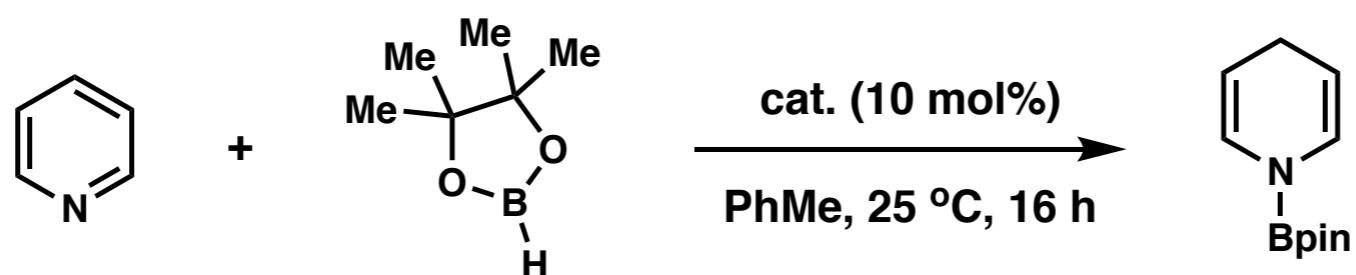
FLPs in Organic Transformations: Borylation

Fontaine, 2015



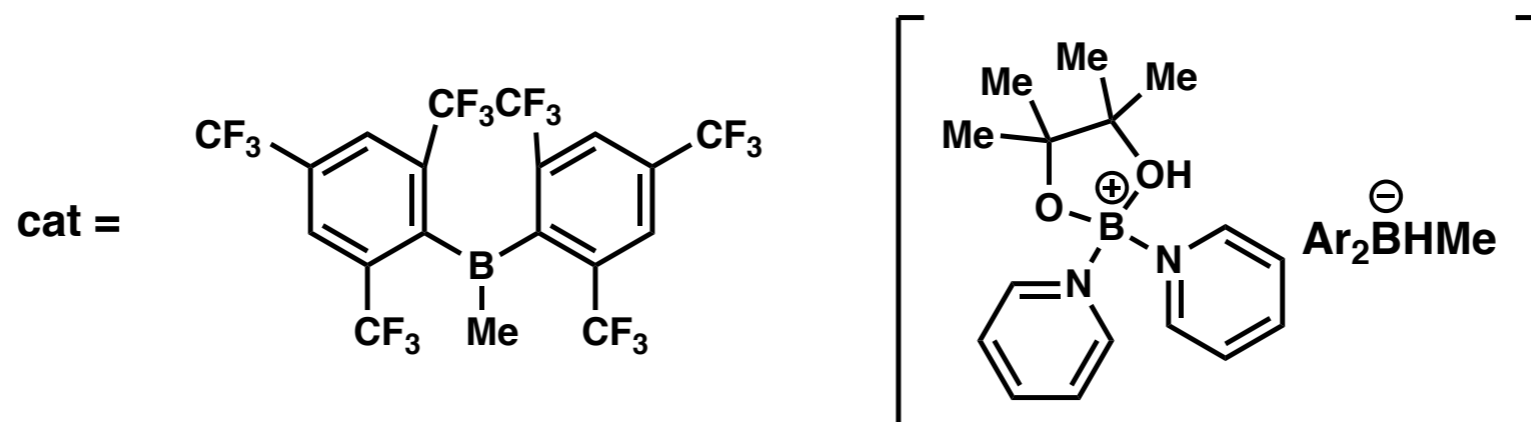
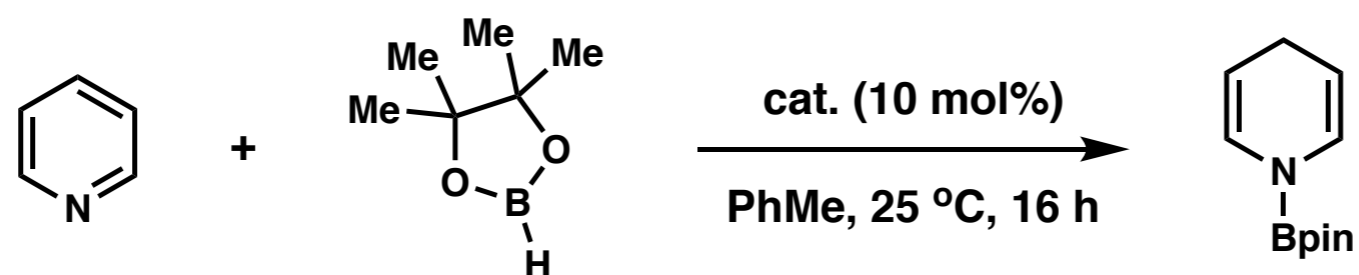
FLPs in Organic Transformations: Hydroboration

Li and Wang, 2015



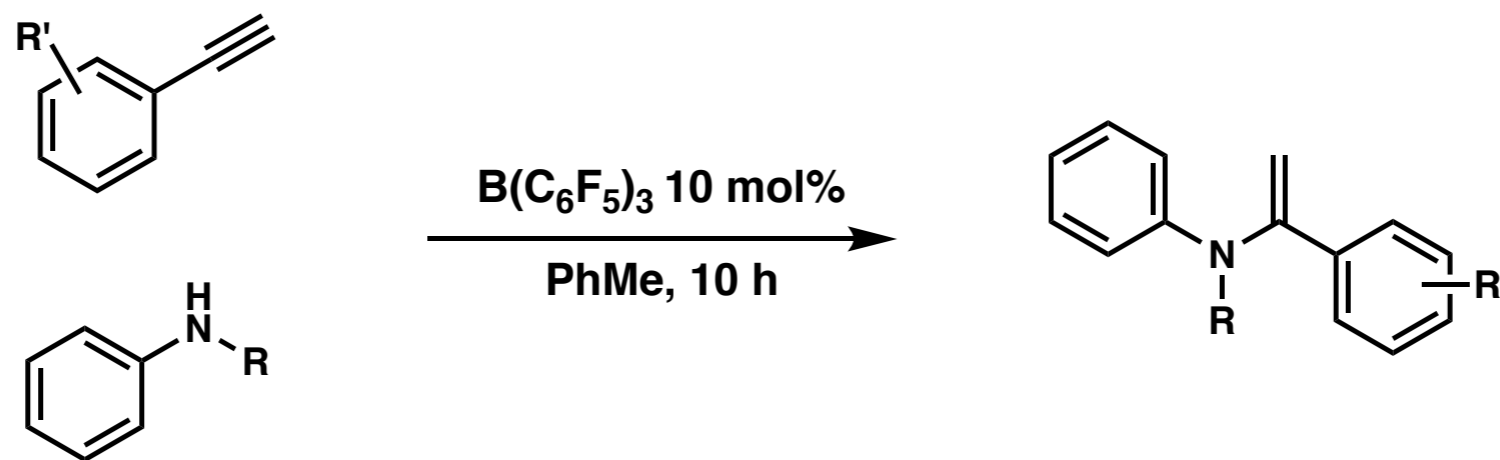
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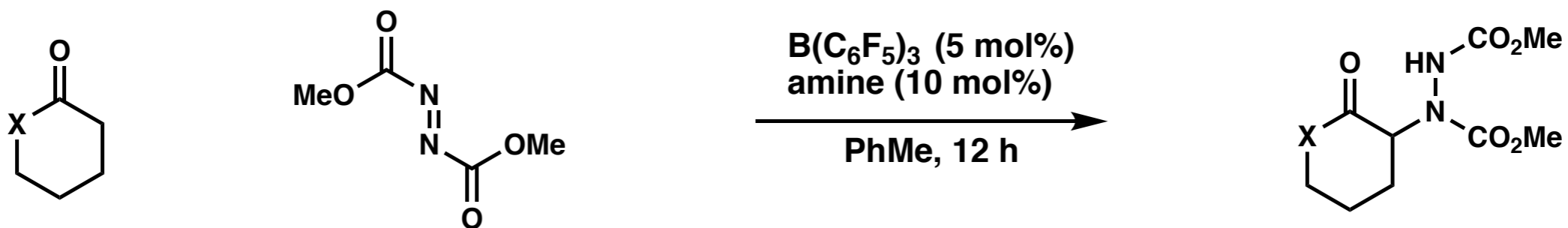
FLPs in Organic Transformations: Hydroamination

Stephan, 2013



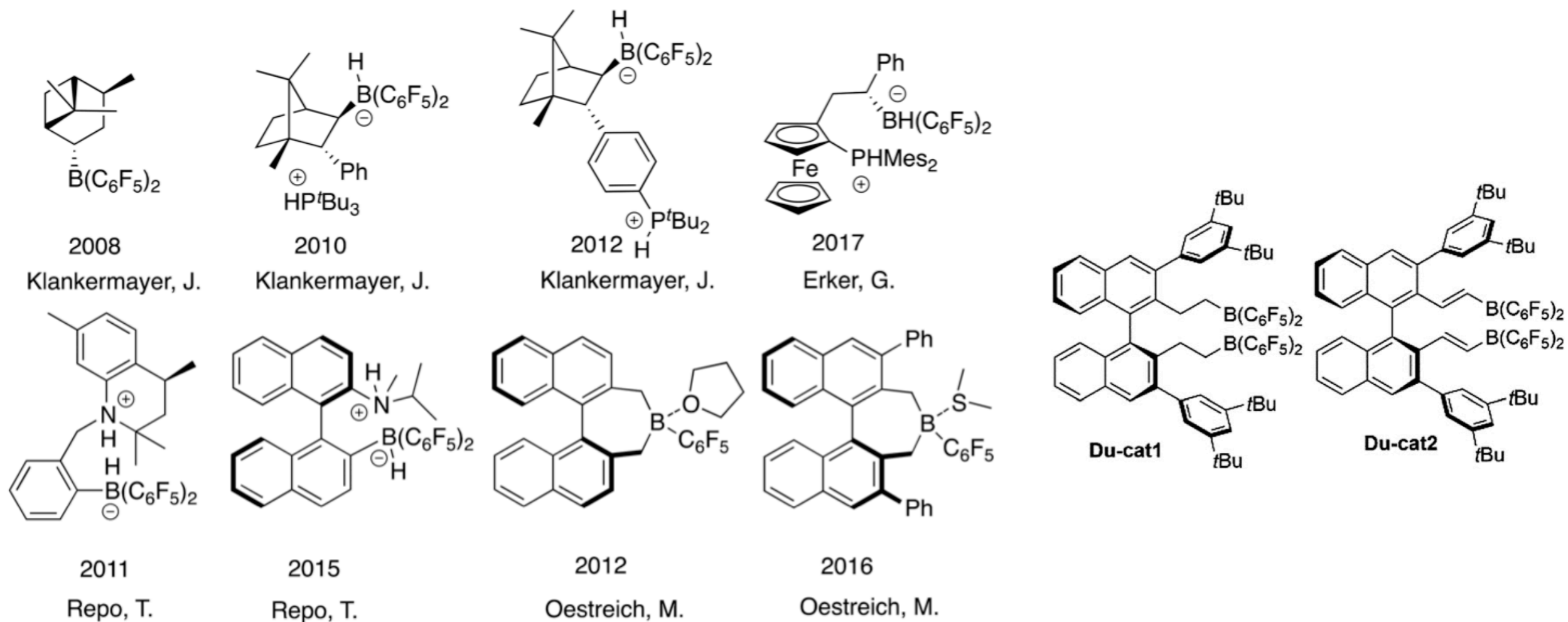
FLPs in Organic Transformations: Amination

Wasa, 2016

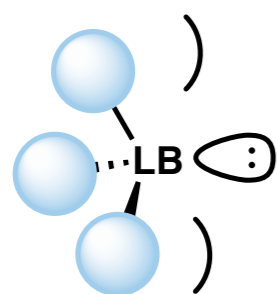


X = CH₂, O, NR, S

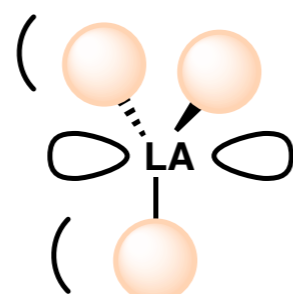
FLPs in Organic Transformations: Chiral Catalysts



Influence of FLP Concept



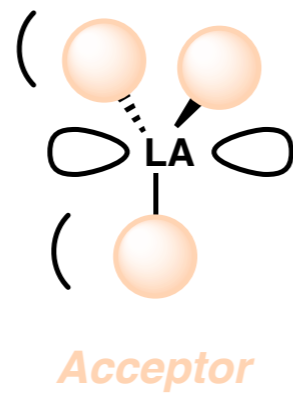
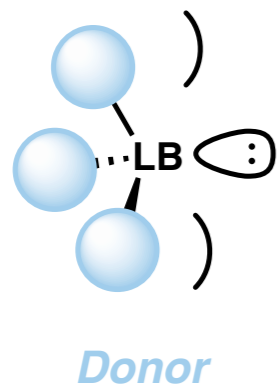
Donor



Acceptor

- **Powerful reactivity platform**
(main group catalysis)

Influence of FLP Concept



- **Powerful reactivity platform**
(main group catalysis)
- **Concept wide reaching**
(enzyme models, polymer chemistry, ligand design for transition metal catalysis, surface chemistry)