Wigner 121
Scientific Symposium

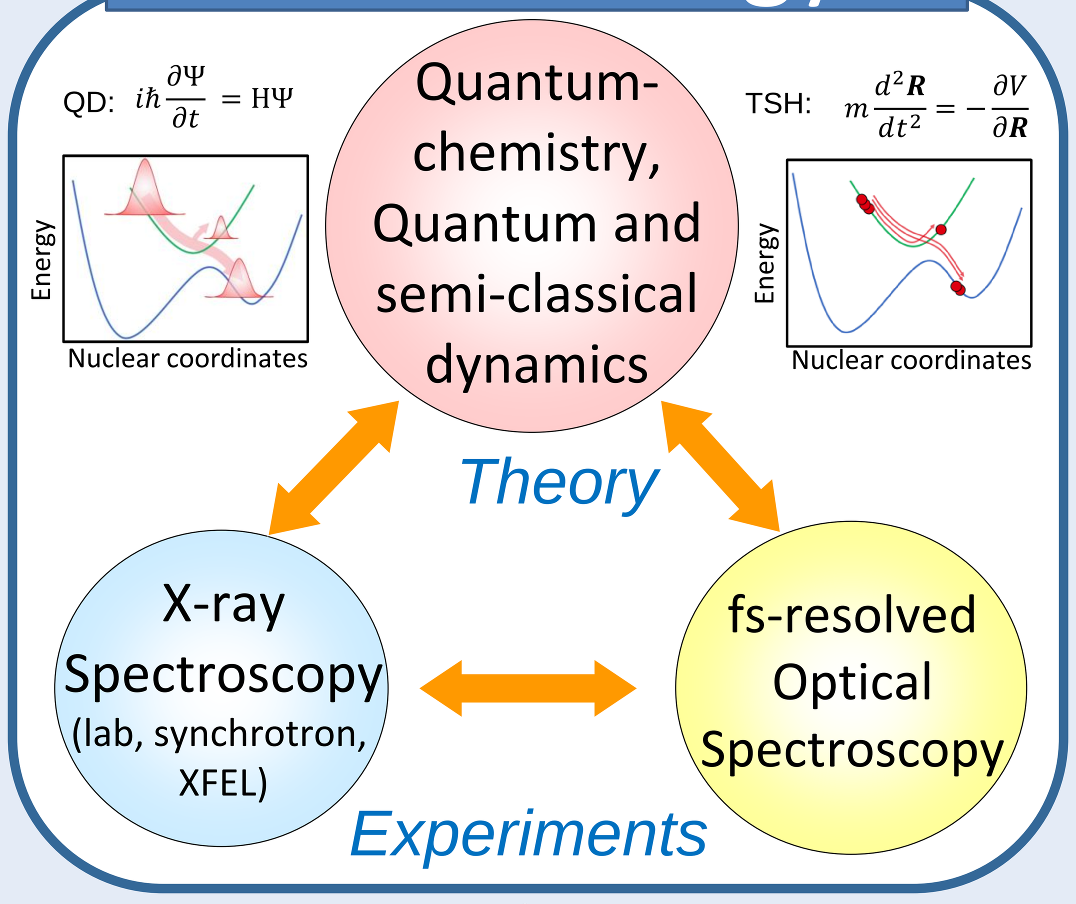
Wigner Research Centre for Physics
Institute for Particle and Nuclear Physics
Femtosecond Spectroscopy and X-ray Spectroscopy Group

G. Vankó , É. G. Bajnóczi, G. Bazsó, D. Buzsáki, J. El Guettoui, T. Keszthelyi, A. J. Mikeházi, Z. Németh, M. Pápai, M. Papp, T. Rozgonyi, A. Vancza

Introduction

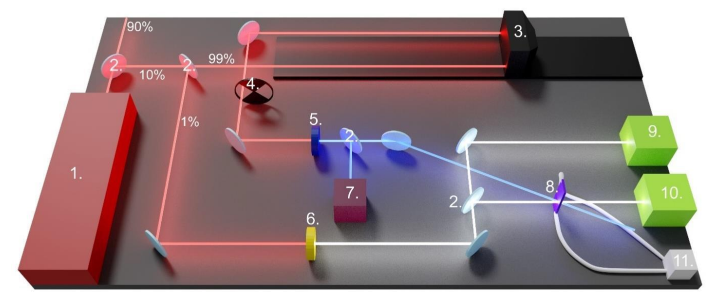
The group has been continuously developing theoretical and experimental tools in order to investigate the ultrafast photoinduced dynamics in functional molecules, and to design new ones with improved performance. The experimental tools include pioneering static high-energy-resolution laboratory X-ray spectroscopy, with hard X-ray absorption being performed routinely and efficiently, while X-ray emission spectroscopy (XES) is still being optimized. Ultrafast studies with X-ray spectroscopy and scattering (XSS) probes are carried out at synchrotron radiation sources and X-ray free electron lasers. In addition, we have realized in our home laboratory a femtosecond-resolved transient optical absorption spectroscopy station, and a femtosecond stimulated Raman scattering setup is being built. Our theoretical efforts focus on designing new functional molecules with quantum chemistry (which we later attempt to synthesize and then subject to experimental tests), as well as running (quantum) dynamics simulations to model the dynamics in light activated molecular systems. Below we show results obtained on a light switchable Fe(II) system.

Methodology



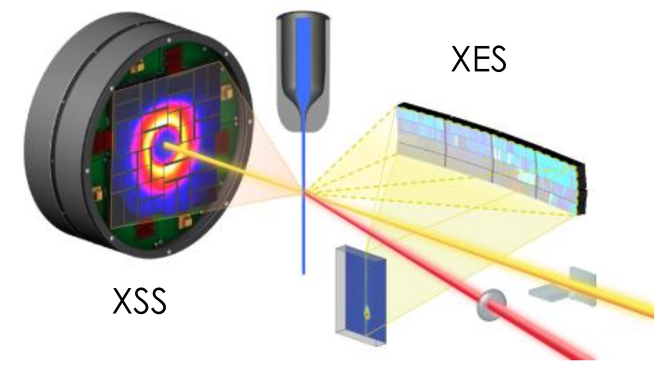
Experiments

Transient Absorption



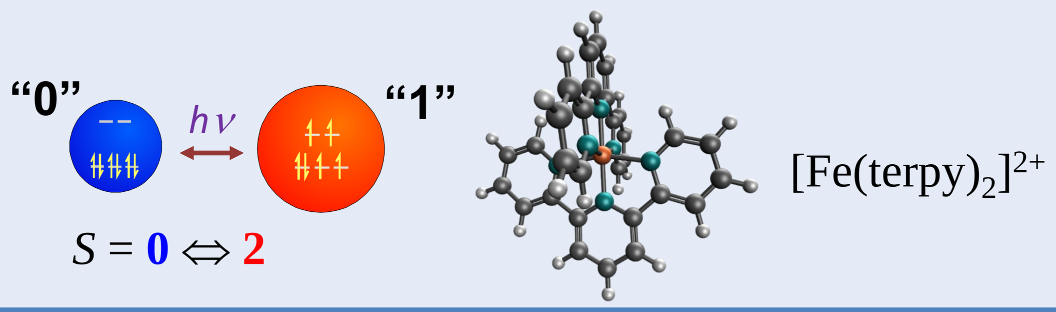
1. Laser source
2. Beamsplitters
3. Linear actuator with retroreflector
4. Beam splitter
5. SHG
6. White light generation
7. Photodiode
8. Flow-through cuvette
9. Reference detector
10. Sample detector
11. Peristaltic pump

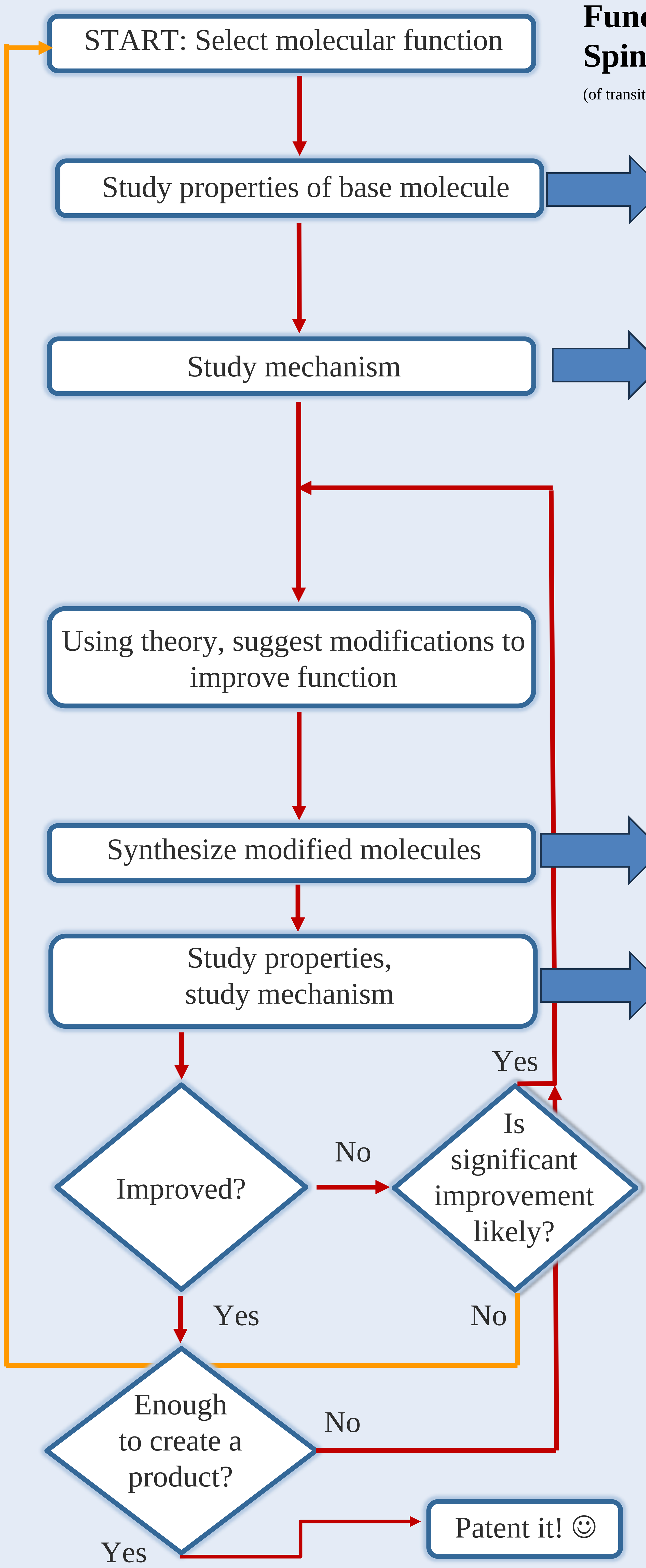
X-ray probes



Function: Spin State Switching

(of transition metal complexes)

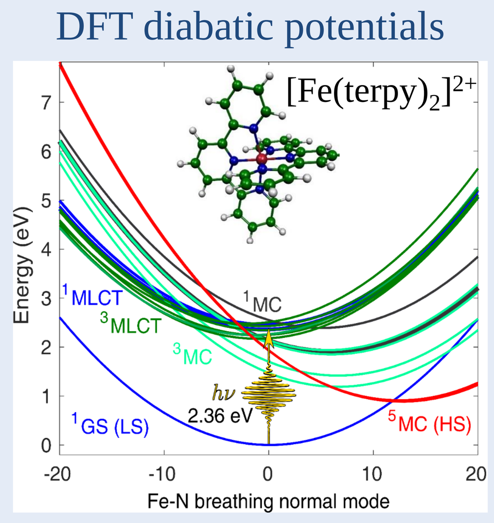




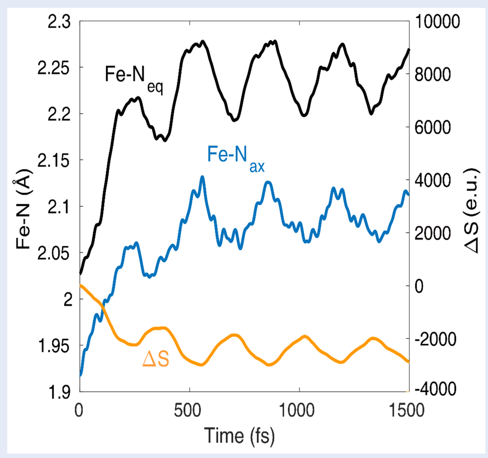
Simulating photo-induced ultrafast excited state dynamics:

$[Fe(terpy)_2]^{2+}$ – Full-D LVC-TSH Dynamics

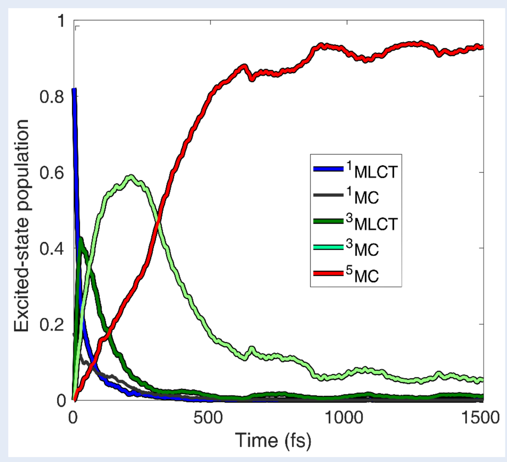
DFT diabatic potentials



Wave-packet dynamics

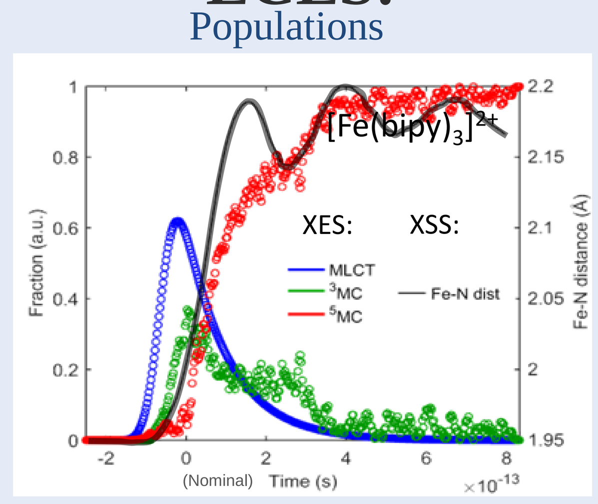


Populations



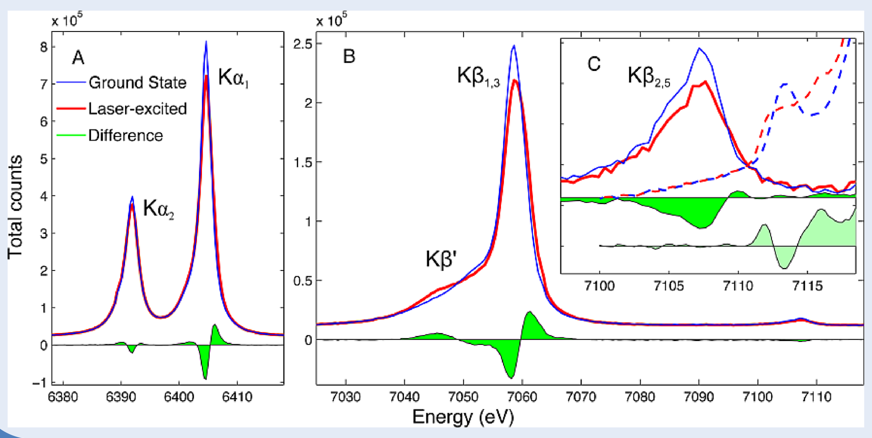
fs-resolved XES/XSS experiments at LCLS:

Populations

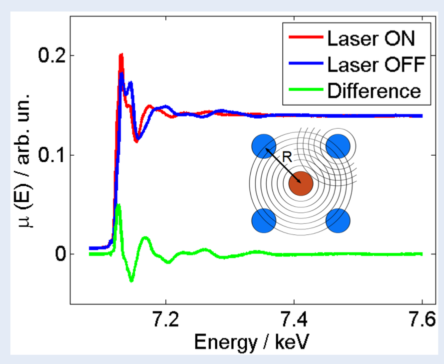


X-ray studies on the quintet excited state of the $[Fe(terpy)_2]^{2+}$ complex:

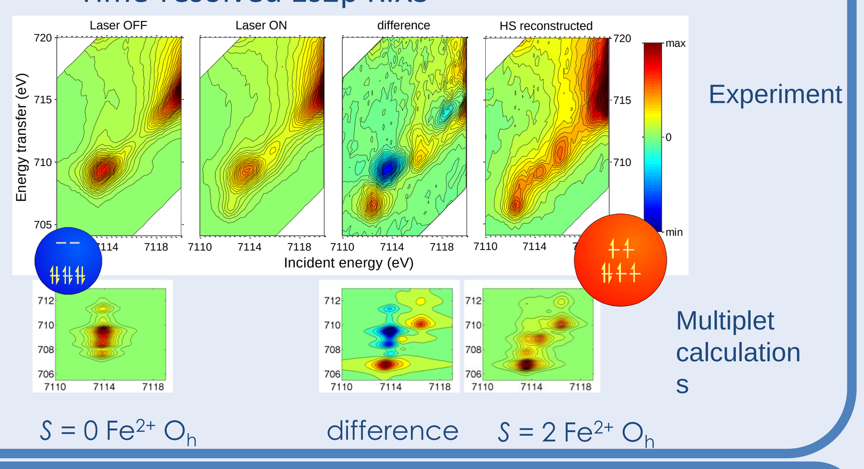
Time-resolved full 1s X-ray emission (XES)



Time-resolved 1s X-ray absorption (XAFS)



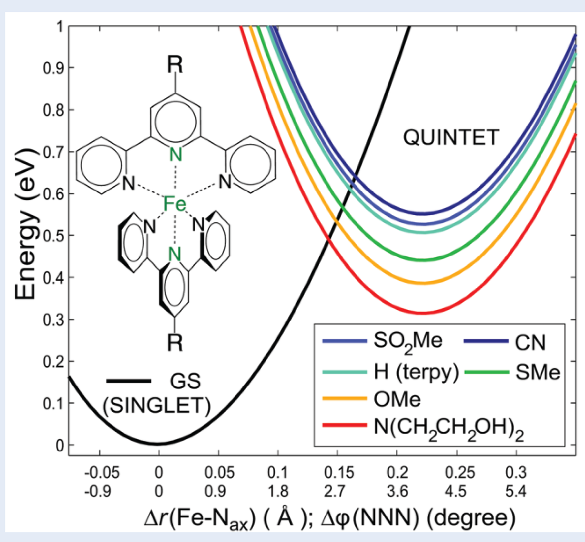
Time-resolved 1s2p RIXS



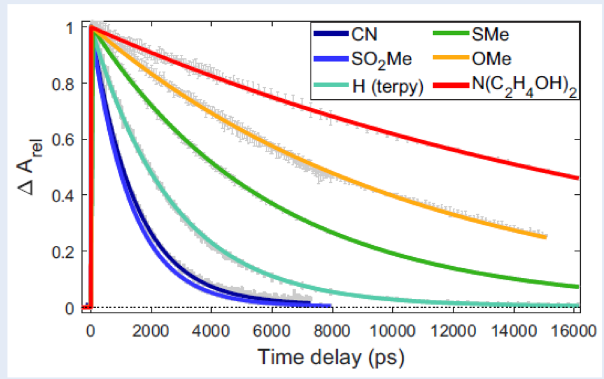
Tailoring properties with substitution

Changing electron density along the molecular axis

Calculated potentials for derivatives



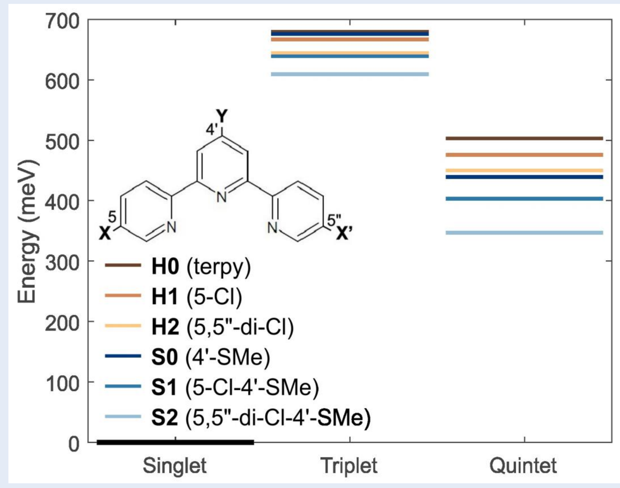
Measured quintet decay



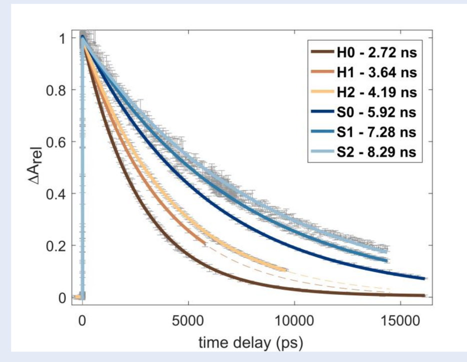
axial electron donation increases quintet lifetime

Changing electron density with side ring substitution

Calculated potentials



Measured quintet decay



equatorial ED also increases quintet lifetime, but less effective

Selected publications of the group

(1) Rozgonyi, T. et al. *Commun Chem* **2023**, *6* (1), 1.
(2) Papp, M. et al. *Inorg. Chem.* **2023**, *62* (16), 6397.
(3) Pápai, M. *J. Chem. Theory Comput.* **2022**, *18* (3), 1329.
(4) Mikeházi, A. et al. *J Synchrotron Rad* **2022**, *29* (5), 1216.
(5) Pápai, M. *Inorg. Chem.* **2021**, *60* (18), 13950.
(6) Kunnus, K. et al. *Nat Commun* **2020**, *11* (1), 634.
(7) March, A. M. et. al. *J. Chem. Phys.* **2019**, *151* (14), 144306.
(8) Britz, A. et al. *Inorg. Chem.* **2019**, *58*, 9341.
(9) Vankó, G. et al., *J. Phys. Chem. C* **2015**, *119*, 5888.
(10) S. Kjær, K. et al. *Phys. Chem. Chem. Phys.* **2018**, *20* 4238.
(11) March, A. M. et. al. *J. Phys. Chem. C* **2017**, *121*, 2620.
(12) Kjaer, K. et al. *Chemical Science* **2019**, *10*, 5749
(13) Katayama, T. et al., *Nat Commun* **2019**, *10*, 3606
(14) Sárosiné Szemes D., *Chem. Commun.* **2020**, *56*, 11831.

HUN-REN Hungarian Research Network

MTA Centre of Excellence