Green Photoredox Catalysis using Near-Infrared Light: Two-Photon-Absorption Enabled Cyclization

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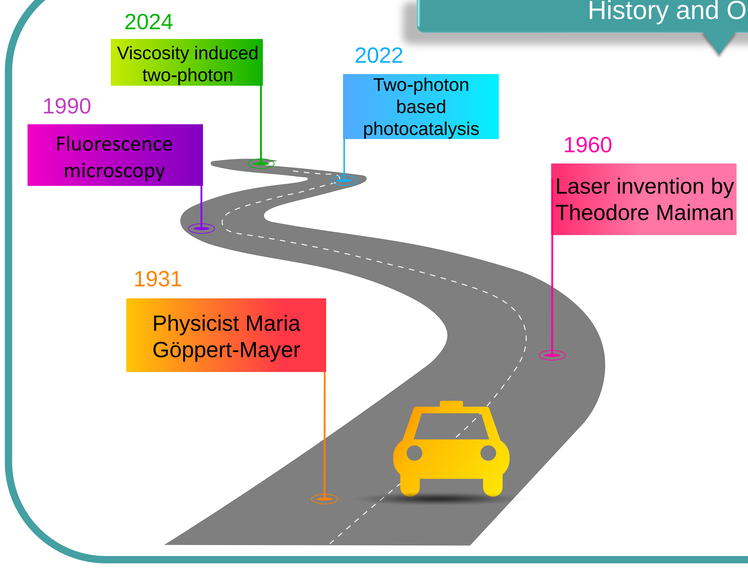
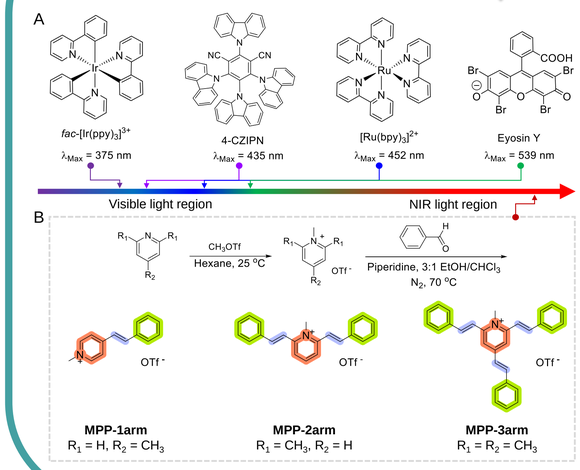
History and Objectives



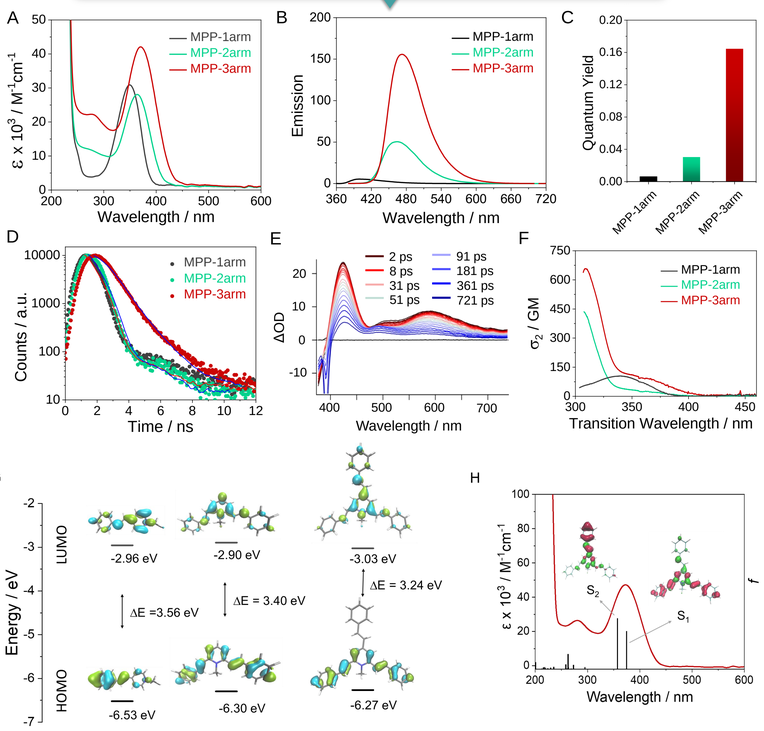
- **1931** Maria Göppert-Mayer, a physicist, first predicted the phenomenon of two-photon absorption (TPA) in her Ph.D. thesis
- **1960** The invention of the laser by Theodore Maiman provided the high-intensity, coherent NLO light necessary to observe TPA
- **1990** Two-photon excitation fluorescence microscopy, a high-resolution imaging technique, was developed by Winfried Denk, James Strickler, and Watt W. Webb.
- **2022** TPA excited photoredox catalysis using highly conjugated Ru(II) photosensitizers (*Nat. Commun.*, **2022**, 13, 2288)
- **2024** Metal-free organic photocatalysts for two-photon-absorption enabled near-infrared light driven benzimidazole cyclization

Background and Syntheses of Photosensitizers



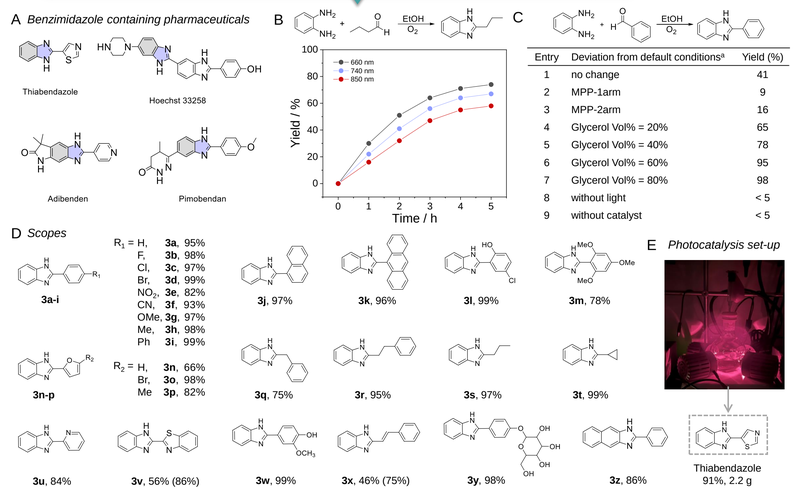
- Metal-free organic photosensitizer for TPA induced NIR light-driven cyclization as green photocatalysis
- Pyridine-based organic photocatalysts with tunable branched donor-acceptor (D-A) structures are designed, which show viscosity-dependent TPA and emission features
- Large TPA cross-section (~600 GM) and long triplet state lifetime, can help to achieve the utilization of NIR light and control the energy transfer for efficient photocatalytic condensation of *o*-phenylenediamine with aldehydes
- Overcoming limitations such as side reaction, low penetration, competing absorption, and incompatibility with light-sensitive substrates

Characterization of Photosensitizers

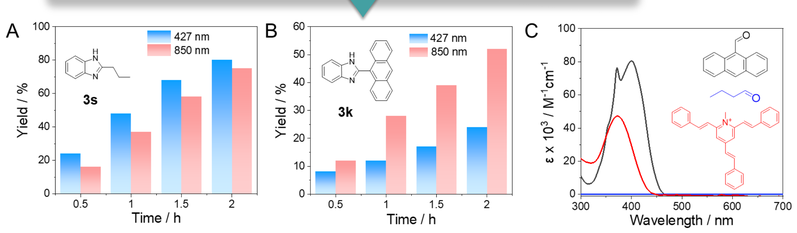


(A) UV-Vis, (B) Fluorescence, (C) Quantum yield (ϕ), (D) Lifetime decay, (E) Transient absorption and (F) Two-photon absorption spectroscopy. (G) DFT and (H) TDDFT / EDDM calculations.

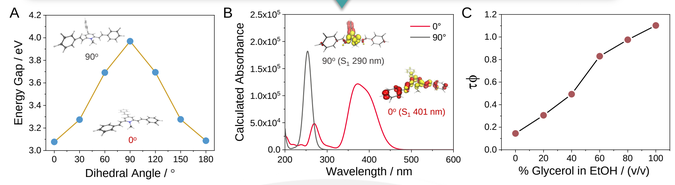
Results and Discussion



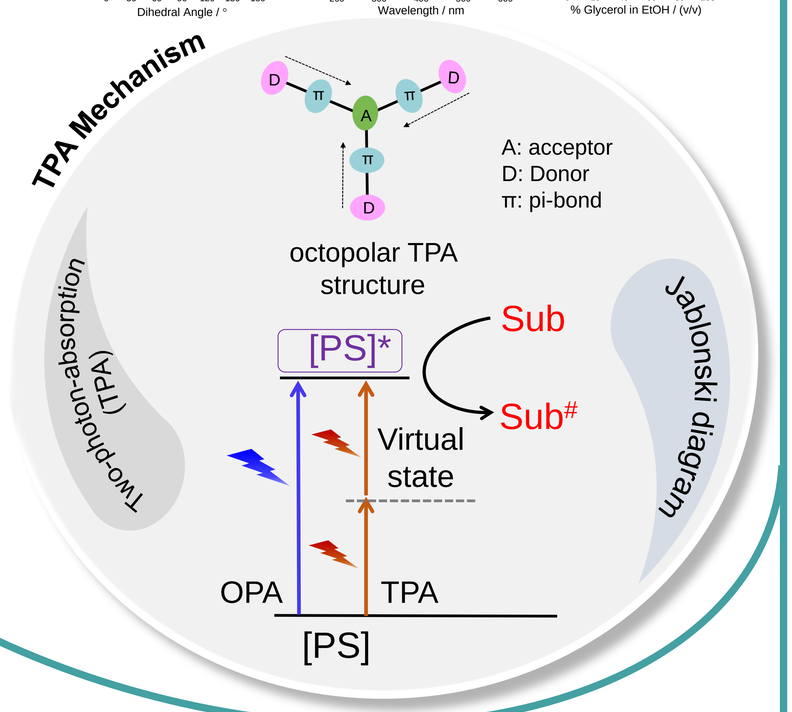
Competing Absorbance Study



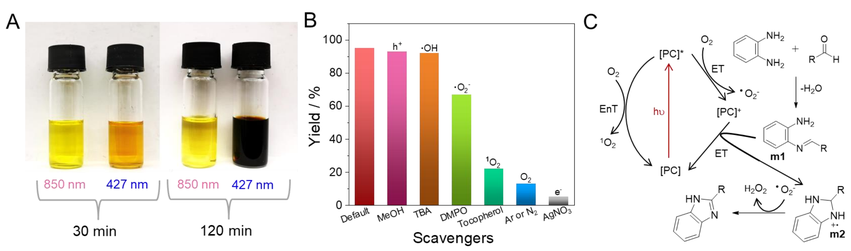
DFT Calculated Dihedral Angles and EDDMs



TPA Mechanism



Conclusion and Future Directions



References and Acknowledgements

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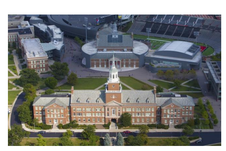

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