

## High-multiplicity states in luminescent organic radicals

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Organic luminescent radicals present a new and exciting platform for exploring molecular functionalities at the interface of their unique optical and spin properties. [1] Near-unity internal quantum efficiency for red emission was demonstrated for donor-acceptor structures based on the tris(2,4,6-trichlorophenyl)methyl (TTM) radical. [2] This opens these materials towards applications in technologies ranging from organic light-emitting diodes and photovoltaics with eliminated triplet losses, to photomagnetic devices and molecular-scale information transfer.

Here we present a new family of molecules based on the TTM radical motif which contain high-multiplicity states in their emission mechanism. By combining light-induced EPR with ultrafast transient absorption and temperature-dependent transient luminescence spectroscopy we find a mechanism that uniquely supports both a high emission yield and a high yield of generation of the high-spin species. This is enabled by very low activation barriers between excited states, which arises from energy level matching in our molecular design. Pulsed EPR spectroscopy at both X and W-band frequencies allows us to characterize the potential of our system for future quantum information science applications.

### References

[1] Teki, Y., *Chem. Eur. J.* **26**, 980 (2020).

[2] Ai, X., Evans, E.W., Dong, S. *et al.*, *Nature* **563**, 536–540 (2018).

