

## Tuning the optical and magnetic properties of photogenerated three-spin systems

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Photogenerated organic triplet-doublet systems hold great promise for a range of technological applications in the emerging field of molecular spintronics, including the areas of quantum information technology and artificial photosynthesis.<sup>[1–4]</sup> These systems are typically composed of a chromophore that is covalently linked to a stable radical. The proximity of the radical alters the excited state dynamics of the chromophore and, frequently, the radical acts as an efficient triplet sensitizer by enhancing the intersystem crossing rate. The further development of any applications involving such systems will crucially depend on our ability to control the magnetic properties of these materials, which requires a profound understanding of the underlying competing excited state deactivation processes. In this contribution, we present our recent results obtained for a range of chromophore–radical compounds comprising systematic modifications of the molecular structure. Perylene diimide (PDI) is used as the chromophore in all cases, but the compounds differ with respect to the number of PDI core substituents, linker type, linker length, and type of the radical. We show that the combination of several complementary spectroscopic techniques, including femtosecond transient UV-vis absorption and transient pulse EPR spectroscopies, allows us to establish a complete picture of the excited state deactivation processes and interactions between the two spin centers. Combining the results from several studies on related structures with insight from ab-initio calculations, we shed light on the factors influencing the rate and yield of triplet formation as well as the magnitude of the exchange interaction between chromophore triplet and radical.

### References

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