Pumping and Pushing Around Electron Spins in Molecular Dyads and Triads

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Molecular dyads and triads are ideal model systems to study the spin evolution in spin correlated radical pairs and its dependence on an external magnetic field. For a series of model systems with a triarylamine electron donor and either naphthalene diimide or perylene diimide acceptor we demonstrate the magnetic field dependence of spin interconversion to be mainly caused by isotropic and anisotropic hyperfine coupling interaction over a magnetic field range up to 2 T.[1-3] Using a new, purely optical pump push spectroscopic method, we could prove the coherent nature of the S-T_0 spin evolution at a variety of fields. In fact, the dominant hyperfine coupling of the triarylamine nitrogen determines the observed quantum beating frequency.[4] For selected examples, also the g-value difference of the two radical centres gains importance at higher fields.

[1] J. H. Klein *et al., J. Am. Chem. Soc.* **137**, 11011 (2015)
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[3] D. Mims *et al., J. Chem. Phys.* **151**, 244308 (2019)
[4] D. Mims *et al.*, *Science* **374**, 1470 (2021)