

Pumping and Pushing Around Electron Spins in Molecular Dyads and Triads

Christoph Lambert (1), Johannes Klein (1), Julian Schäfer (1), Stefan Riese (1), David Mims (1), Paul Metzel (1), Chantal Roger (1), Nikita Lukzen (2), Ulrich Steiner (3)

(1) Institute of Organic Chemistry, University of Würzburg,

Am Hubland, 97074 Würzburg, Germany

christoph.lambert@uni-wuerzburg.de

(2) International Tomography Center, Institutskaya 3a,

and Novosibirsk State University, Novosibirsk 630090, Russia

(3) Department of Chemistry, University of Konstanz,

Universitätsstraße 14, 78457 Konstanz, Germany

ulrich.steiner@uni-konstanz.de

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-T₀ 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 and/or the g-tensor anisotropy of the two radical centres gains importance at higher fields.

[1] J. H. Klein *et al.*, *J. Am. Chem. Soc.* **137**, 11011 (2015)

[2] J. Schäfer *et al.*, *Phys. Chem. Chem. Phys.* **20**, 27093 (2018)

[3] D. Mims *et al.*, *J. Chem. Phys.* **151**, 244308 (2019)

[4] D. Mims *et al.*, *Science* **374**, 1470 (2021)