

Spin-Spin Interactions and their Impact on Organic Light-Emitting Devices

Vladimir Dyakonov, Andreas Sperlich

Julius Maximilian University of Würzburg, 97074 Würzburg, Germany

vladimir.dyakonov@uni-wuerzburg.de

Spin-spin interactions in organic light-emitting diodes (OLEDs) based on thermally activated delayed fluorescence (TADF) are pivotal because radiative recombination is largely determined by triplet-to-singlet conversion, also called reverse intersystem crossing (RISC). To explore the underlying process, we apply a spin-resonance spectral hole-burning technique to probe electroluminescence. We find that the triplet exciplex states in OLEDs are highly spin-polarized and show that these states can be decoupled from the heterogeneous nuclear environment as a source of spin dephasing and can even be coherently manipulated on a spin-spin relaxation time scale T_2^* of 30 ns. Crucially, we obtain the characteristic triplet exciplex spin-lattice relaxation time T_1 in the range of 50 μ s, which far exceeds the RISC time. We conclude that slow spin relaxation rather than RISC is an efficiency-limiting step for intermolecular donor:acceptor systems. Finding TADF emitters with faster spin relaxation will benefit this type of TADF OLEDs. [1].

References

[1] S. Weissenseel *et al.*, *Science Adv.* **7**, abj9961 (2021).