Room temperature radical-pair spin relaxation dynamics at low magnetic fields studied by spin-dependent charge carrier recombination currents in organic light-emitting diodes

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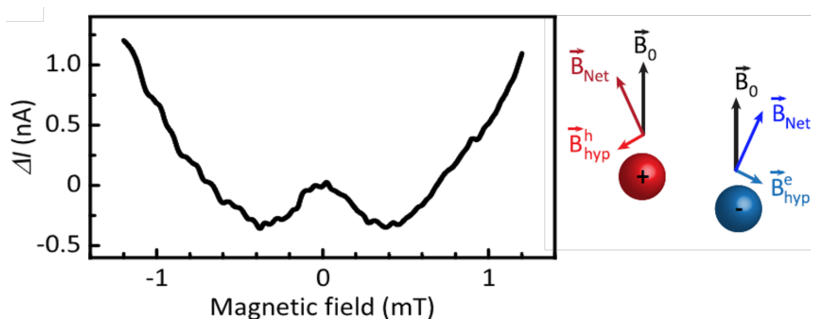
We have experimentally tested the hypothesis that the strong magnetic field dependence of radical-pair-like processes is related to a strong magnetic field dependence of spin-relaxation times when an applied magnetic field competes in magnitude with internal, proton-hyperfine induced magnetic fields. Electric current in bipolar (electron/hole) injector devices, essentially organic light-emitting diodes (OLEDs) under forward bias, provides straightforward experimental access to spin-dependent charge carrier recombination rates, which have been known to be qualitative analogues to spin-dependent radical pair reaction rates[1]. We used such spin-dependent electric currents to observe pulsed electrically detected magnetic resonance, specifically electrically detected Hahn-echos[2] for the measurement of charge carrier spin coherence times T_2; and electrically detected inversion recovery[2] for the measurement of longitudinal charge carrier spin relaxation times T_1. These measurements were performed in a regime, where the static magnetic field (B_0) is so small that magnetic polarization is negligible (1 mT $\lesssim B_0 \lesssim$ 8 mT)[3]. The experiments required arbitrary waveform generation (AWG) for the direct synthesis of the RF pulse sequences needed for coherent spin-control. The results of this study have revealed a strong magnetic-field dependence of T_1 at magnetic field strengths where radical-pair processes, e.g. magnetoresistance, are particularly magnetosensitive. In conclusion, we see that when B_0 becomes so small that it is essentially cancelled by the randomly oriented hyperfine fields within the thin-film material, the individual spin pairs lose their well-defined quantization axis and a T_1 process is not well-defined anymore[3]. Measurements of T_1, therefore, reveal values that are strongly quenched, converging towards the value of T_2.



Room temperature current change in an OLED based on the π-conjugated co-polymer SY-PPV, under forward bias conditions, as a function of an applied mT-range magnetic field. The sketch illustrates how an externally applied magnetic field and locally varying hyperfine field components add up to the net magnetic field that acts upon individual charge carriers. When these net fields are dominated by B_0, a well-define identical quantization axis will exist for all charge carrier spin pairs within the polymer film.

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[1] T. Grünbaum et al., *Faraday Discuss.* **221**, 92 (2020).
[2] W. J. Baker et al., Phys. Rev. Lett. **108**, 267601 (2012).
[3] T. H. Tennahewa et al., arXiv:2207.07086 [cond-mat.mes-hall] (2022).