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 rates¹. We used such spin-dependent electric currents to observe pulsed electrically detected magnetic resonance, specifically electrically detected Hahn-echoes² for the measurement of charge carrier spin coherence times $T_2$; and electrically detected inversion recovery² 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 ($1 \text{ mT} \lesssim B_0 \lesssim 8 \text{ mT}$)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 anymore³. Measurements of $T_1$, therefore, reveal values that are strongly quenched, converging towards the value of $T_2$.

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