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### **Spin-dependent electronic processes in organic semiconductors<sup>1</sup>**

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The development and improvement of organic electronics and spintronics concepts [1, 2] requires a detailed understanding of spin-dependent charge-carrier transitions, including transport and recombination processes, which govern the magneto-optoelectronic properties of these materials. In order to observe these processes, we developed various electrically detected magnetic resonance (EDMR) experiments, in particular pulsed EDMR experiments and we have applied these techniques to a range of polymer materials and devices. EDMR allows for the observation of spin-dependent electronic rates after the spin manifolds controlling these processes have been excited by short and powerful microwave pulses. Most polymers typically exhibit small, but non-zero spin-orbit coupling effect in their EDMR spectra due to the absence of heavy elements. They also display abundant hyperfine coupling between charge-carrier and adjacent nuclear spins, which are abundant in most organic materials [3]. While spectroscopically, the inhomogeneous broadening effects of EDMR lines by these two coupling types is indistinguishable (both display Gaussian lines due to the all abundant magnetic disorder), they can be separated when EDMR experiments are conducted over a wide range of different excitation frequencies, ranging from the radio-frequency domain where spin-orbit coupling is negligible and electronic transitions are predominantly governed by the weak random hyperfine fields, to the quasi-optical domain where differences and anisotropies in the charge-carrier g-factors emerge [4]. EDMR is uniquely suitable for experiments at very low excitation frequencies where spin polarization is negligible. This facilitates the study of spin collectivity in an ultra-strong coupling regime under strong radiofrequency drive [5, 6] as well as calibration-free magnetic field sensor applications that utilize magnetic resonance [2]. [1] Xiong et al., Nature 427, 821-824 (2004). [2] Baker et al., Nat. Commun. 3, 898 (2012). [3] Malissa et al., Science 345, 1487-1490 (2014). [4] Joshi et al., Appl. Phys. Lett. 109, 103303 (2016). [5] Roundy and Raikh, Phys. Rev. B 88, 125206 (2013). [6] Waters et al., Nat. Phys. 11, 910 (2015).

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