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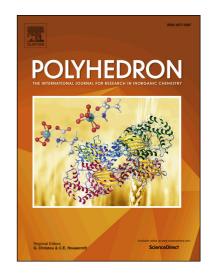
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## **ACCEPTED MANUSCRIPT**

Probing electron-hole pairs in polymer light emitting diodes using electrically- and electroluminescence- detected magnetic resonance techniques

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Abstract. Despite the significance of weakly coupled electron-hole (e-h) pairs in considering operation mechanisms of organic light emitting diode (OLED) and magnetic field effects (MFEs), behaviors of e-h pairs in operating OLEDs have been experimentally unexplored. Electrically detected magnetic resonance (EDMR) and electroluminescence detected magnetic resonance (ELDMR) techniques have been applied to polymer OLEDs to probe the e-h pairs in operating OLEDs. The spectra of EDMR and ELDMR resemble each other, suggesting that e-h pairs are responsible for EDMR and ELDMR signals. Both signals are found to stem from equilibrium shifts of singlet e-h pairs (SPs) toward triplet e-h pairs by the ESR transition. We show that the voltage dependence of EL- and ELDMR- intensities are similar, indicating that the voltage dependence of the SP-density can be predicted from that of the ELDMR-response in the SP-density.

#### 1. Introduction

Organic semiconductors have attracted much interest due to their potential applications to thin-film devices such as organic light emitting diodes (OLEDs) and solar cells. In particular, highly efficient and thin displays based on OLEDs have already been in commercial production. In normal operation of fluorescent OLEDs, electrons and holes combine to form excitons and emission of light (electroluminescence: EL) occurs by the transition of singlet excitons (SEs) to the ground state. Electron-hole (e-h) pairs then generate as intermediate states between free

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