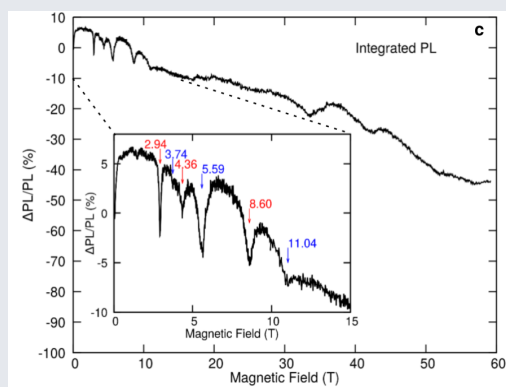


# Site-selective measurement of coupled spin pairs in an organic semiconductor

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From organic electronics to biological systems, understanding the role of intermolecular interactions between spin pairs is a key challenge. We showed that such pairs can be selectively addressed using a combination of experiments at high magnetic fields (up to 60 Tesla) and optical spectroscopy. We demonstrated this for bound pairs of spin-triplet excitons formed by singlet fission, but our approach has direct applicability to a wide range of synthetic and biological systems. To address the triplet pairs in a site-sensitive way, we tuned the magnetic field to match the site dependent exchange energy of the pair, creating a level crossing resonance between optically bright singlet ( $S=0$ ) and dark triplet, quintet ( $S=1,2$ ) configurations. This induces narrow holes in a broad optical emission spectrum, uncovering exchange-specific luminescence. We identified three distinct triplet-pair sites, with exchange couplings varying over an order of magnitude (0.3-5 meV), each with its own luminescence spectrum, coexisting in a single material. Our results reveal how site-selectivity can be achieved for organic spin pairs in a broad range of systems



*Photoluminescence in TIPS-tetracene at 2K as function of the magnetic field. Several resonances with characteristic magnetic fields from 3 to 30 Tesla are seen which correspond to crossing between triplet/quintet and singlet states.*

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