

# SPIN PROPERTIES OF BI-EXCITON STATE FORMED THROUGH SINGLET FISSION

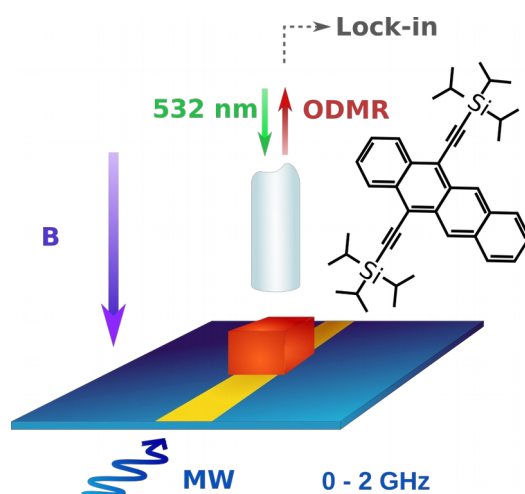
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## ABSTRACT

From organic electronics to biological systems, understanding the role of intermolecular interactions between spin pairs is a key challenge. Here we show how such pairs can be selectively addressed with combined spin and optical sensitivity. We demonstrate this for bound pairs of spin-triplet excitations formed by singlet fission, with direct applicability across a wide range of synthetic and biological systems. We show that the site-sensitivity of exchange coupling allows distinct triplet pairs to be resonantly addressed at different magnetic fields, tuning them 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. Using fields up to 60 T, we identify 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. We then show how broadband optically detected magnetic resonance can allow to find the microscopic positions of the triplet exciton that for a strongly bound quintet state within the crystal structure of TIPS-Tetracene.



## REFERENCES

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[2] Sam L. Bayliss, Alexei D. Chepelianskii, Alessandro Sepe, Brian J. Walker, Bruno Ehrler, Matthew J. Bruzek, John E. Anthony, and Neil C. Greenham, Phys. Rev. Lett. **112**, 238701 (2014)