Triplet-Triplet Annihilation: Magnetic Field Effects in Solution

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In triplet-triplet annihilation (TTA) the molecular energy of two photons is pooled and emitted as fluorescence of a single photon of higher energy. TTA is a promising means of accessing solar irradiance below the silicon bandgap and surpassing the Shockley-Queisser limit. In addition, TTA allows the output light wavelength to be tailored to a specific application via choice of molecule and functionalisation. As TTA is a spin-selective process it exhibits a magnetic field response, which has tradi- tionally been described and modelled in the context of Atkins and Evans' Theory [1].

Here, we revisit the theory, motivating the origin of key equations and evaluating the assumptions behind them. We re-derive the theory with an alternative choice of initial and equilibrium states which better captures the typical situation for TTA in solution. In doing so we also compute the relative contributions of all spin channels, not just the triplet channel. These new conditions fundamentally change the evolution of decoherence in the system, and thus the final magnetic field response equations. The ramifications of these updates are discussed in light of recent experimental results [2].

[1] P. Atkins and G. Evans, Magnetic field effects on chemiluminescent fluid solutions. *Molecular Physics*, 29, 921-935 (1975).

[2] E. M. Gholizadeh, S. K. K. Prasad, Z.L. Teh, T. Ishwara, S. Norman, A. J. Petty, J. H. Cole, S. Cheong, R. D. Tilley, J. E. Anthony, S. Huang, T. W. Schmidt, Photochemical upconversion of near-infrared light from below the silicon bandgap, *Nature Photonics*, 14, 585-590 (2020).