Theory of Spin Effects in Triplet–Triplet Annihilation Upconversion: Modelling Magneto–Photoluminescence

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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 thermodynamic limit for single-junction solar cells. 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 traditionally been described and modelled in the context of Atkins & Evans' Theory.[1] Here, we revisit the theory, motivating the origin of key equations and evaluating the assumptions behind them. We rederive the theory, which captures the typical situation for TTA in solution. We compute the relative contributions of all spin channels,[2] not only the singlet as in the original, but also triplet and quintet channels. These new conditions 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 magneto-photoluminescence experimental results.[3,4]

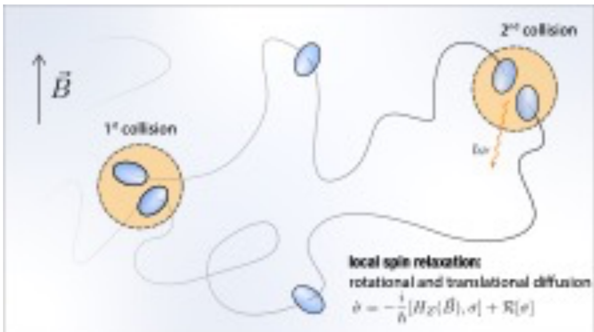

Figure 1: Relaxed triplets hosted on two molecules undergo a first encounter, resulting in energy exchange. The two spins evolve under the effect of the Zeeman Hamiltonian and the stochastic rotational/translational diffusion of the host molecule. This induces a local spin relaxation. The triplets undergo a second collision. The probability of upconversion (emission of a photon) depends on the strength of the magnetic field and the roto-translational parameters.

References
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