

Interaction of a photogenerated spin qubit pair with a stable radical in DNA hairpins

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Tunable molecular systems that can generate entangled electrons, or spin qubit pairs (SQPs), are highly desirable for developments in quantum information science (QIS). Photogenerated SQPs can be initialized in a pure quantum state which exhibits relatively long coherence times, allowing for electron spin manipulation via optical and magnetic spectroscopy. DNA hairpins serve as a modifiable scaffold on which to study these multi-spin interactions, such as a SQP in conjunction with a stable radical. Such a multi-spin system is optimal for implementing spin manipulations that could be advantageous for executing quantum logic gates and spin teleportation. DNA hairpins also offer the possibility of individual qubit addressability and qubit scalability. Here, we examine a series of DNA hairpins that use naphthalenediimide (NDI) as the hairpin linker and chromophore, and guanine or an organic endcap as the terminal hole acceptor. A stable nitroxide radical is covalently attached to the hairpin to facilitate spin interactions between the SQP and stable radical. Electron paramagnetic resonance spectroscopy and ultrafast optical spectroscopy are utilized to explore the charge transfer rates and spin-spin interactions in these DNA hairpins to determine how the SQP pair interacts with the stable nitroxide radical, and how this modulates the overall dynamics of the system.