

Coherences of Photo-Induced Electron Spin Qubit Pair States in Natural Photosynthetic Proteins

Jasleen K Bindra,¹ Jens Niklas,¹ Yeonjun Jeong,¹ Ahren W. Jasper,¹ Lisa M. Utschig,¹ and Oleg G. Poluektov¹

¹*Chemical Sciences and Engineering Division, Argonne National Laboratory, Lemont, IL 60439, USA, J.Bindra@anl.gov*

Photosynthetic proteins represent well-defined and experimentally tunable molecular systems, exhibiting complexities inspired by their functional roles. Due to these characteristics, they serve as ideal model systems for investigating spin coherences. The objective of this study is to unravel how nature manages coherence and spin entanglement in photosynthesis. Despite their significance, critical aspects, like coherence spatial lengths, lifetime, dephasing, decoherence mechanisms, and their interaction with the local and global protein structure, remain poorly understood, hindering a detailed understanding of decoherence in this context. This work presents the first comprehensive experimental study on decoherences in photoinduced electron spin states, in both Type II and Type I reaction centers. High-frequency electron paramagnetic resonance (EPR) spectroscopy operating at 130 GHz and 4.6 T was used to measure coherences through the decay of two-pulse electron spin echo signals and Rabi oscillations. The phase memory times (TM) recorded at various temperatures show that TM exhibits minimal dependence on biological species, biochemical treatment, and paramagnetic species. Nuclear spin diffusion and instantaneous diffusion mechanisms alone cannot explain the observed decoherence. Instead, the low-temperature dynamics of methyl groups surrounding the unpaired electron spin centers are suggested as the main factor governing loss of coherence. Understanding these intricate dynamics holds the key to enhancing our comprehension of photosynthetic processes and their potential applications in achieving more efficient solar energy conversion.

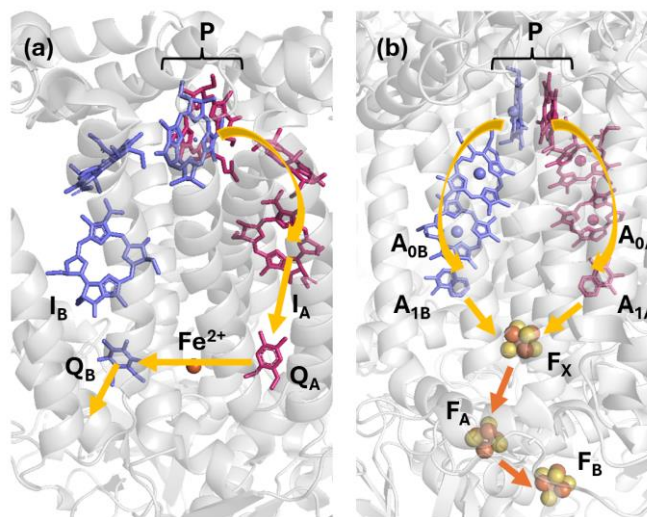


Figure 1. Figure 1: Schematic structure and ET pathways in photosynthetic RCs of Type II (a) and Type I (b).