Supporting Information

Light-Induced Electron Spin Qubit Coherences in the Purple Bacteria Reaction Center Protein

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S9	**Figure S8**	The functional group contribution to the coherence time for the quinone electron spin for bRC (2j8c-U10M) and PSI (1jbo-PQN), as described as the coherence time increase ΔT_M resulting from the removal of the functional group. The symbols indicate the groups: $-CH_3$ (blue squares), $>CH_2$ (red pentagons), $-NH_2$ (green triangles), and H_2O (cyan circles). Protons not assigned to any of these groups are shown as black crosses (x). The calculated values of ΔT_M are plotted as a function the distance from the electron spin, *r*. The overlaid orange line is the coherence time calculated with all protons at distances greater than *r* removed. The magnetic field directions shown are: (a) Q_A, $\hat{g}_y$ for bRC, (b) A_{1A}^-, $\hat{g}_y$ for PSI, (c) Q_A, $\hat{g}_z$ for bRC, and (d) A_{1A}^-, $\hat{g}_z$ for PSI. Results for the x direction are presented in the main article.
S10	**Table S1**	Calculated NSD coherence times T_M for the quinone electron spin for protonated and deuterated bRC. The unmodified results may be compared with the results of artificial calculations where the interactions of the protons or deuterons on the methyl groups were increased by factors of 10–100.

D-band (130 GHz) pulsed EPR spectra of PSI complexes from fully deuterated Zn bRC. The sample was frozen in the dark and cooled down to 40 K and then illuminated. Field-swept P_{865}^{+} Q_A^- radical pair (SCRP) spectra were recorded at different temperatures with 700 ns delay after flash (DAF) and 532 nm Laser light. Laser repetition rate is 20 Hertz (50 ms). The so-called 'dark' and 'light' signals were recorded.

The 'dark' signal is recorded at the midtime point between two Laser pulses. At this point in time not all photoexcited bRCs have experienced charge recombination, but the radical pair (SCRP) has in the meantime thermalized (achieved Boltzmann distribution), since the spin lattice relaxation time T_1 of the electron spins is significantly shorter ($\leq$ 1 ms) than the radical pair recombination time in this temperature range. For this reason, the 'dark' signal is fully absorptive and has no emissive signals. Note, that this signal is only observed at high-frequency EPR due to the much larger thermal polarization (D-band/X-band ~14x). All electron spin pair coherences have long decayed. Subtraction of the this 'dark' signal from the signal directly after the Laser pulse ('light') provides the pure SCRP spectrum ('light-minus-dark').

Figure S1(a) shows dark, (b) light and (c) difference (light-dark). Figure S1(c) shows two low field components correspond to two main components g_x and g_y of the Q_A^- g-tensor. The high field component corresponding to the overlap of the three g-tensor components of P_{865}^{+} and a third low intensity g_z component of Q_A^-. Similar temperature dependence was also obtained for other samples. The recorded field-swept spectra are shown in Figures S1-S3.

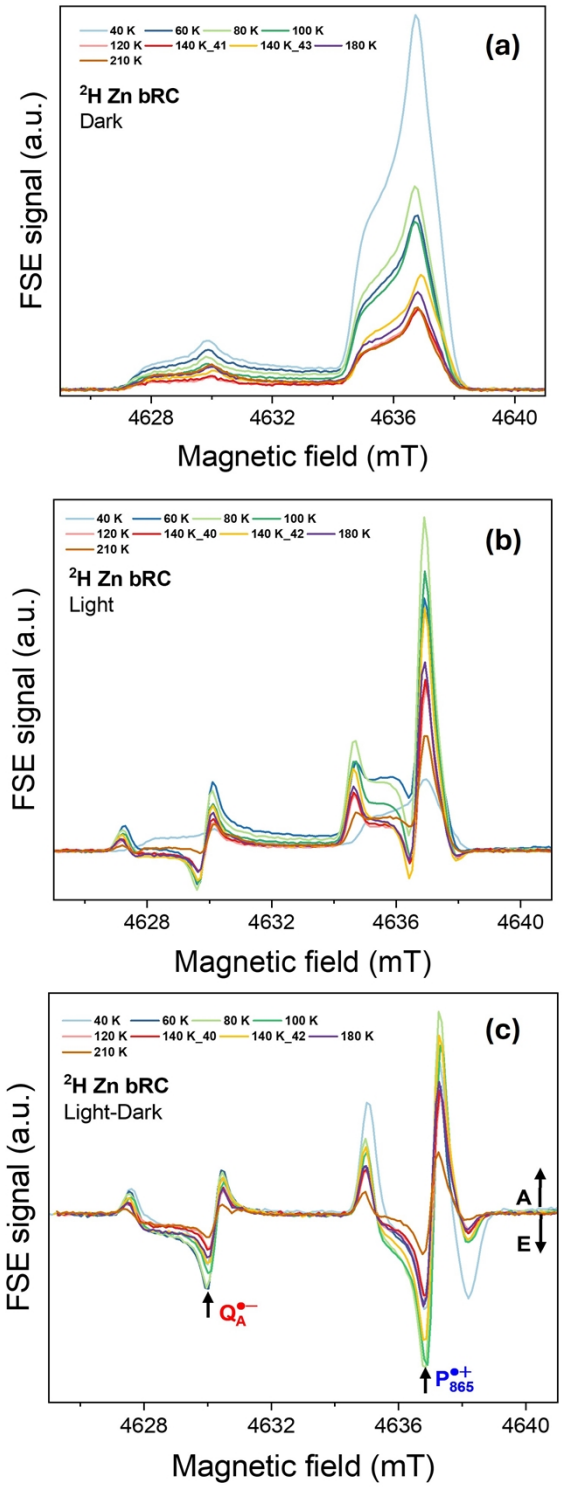

Figure S1: Temperature dependence of field-swept echo detected D-band (130 GHz) EPR spectra "light" (a), "dark" (b) and difference/light-dark (c) for ^{2}H Zn bRC.

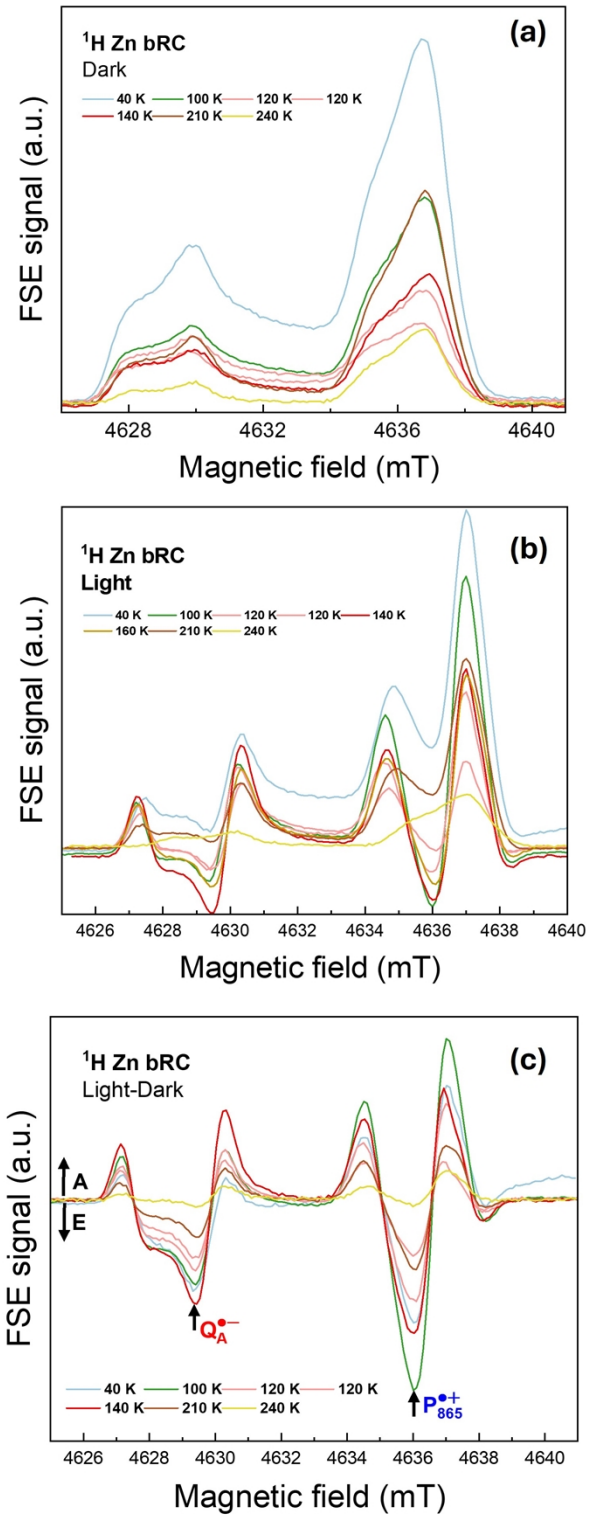

Figure S2: Temperature dependence of field-swept echo detected D-band (130 GHz) EPR spectra "light" (a), "dark" (b) and difference/light-dark (c) for ^{1}H Zn bRC.

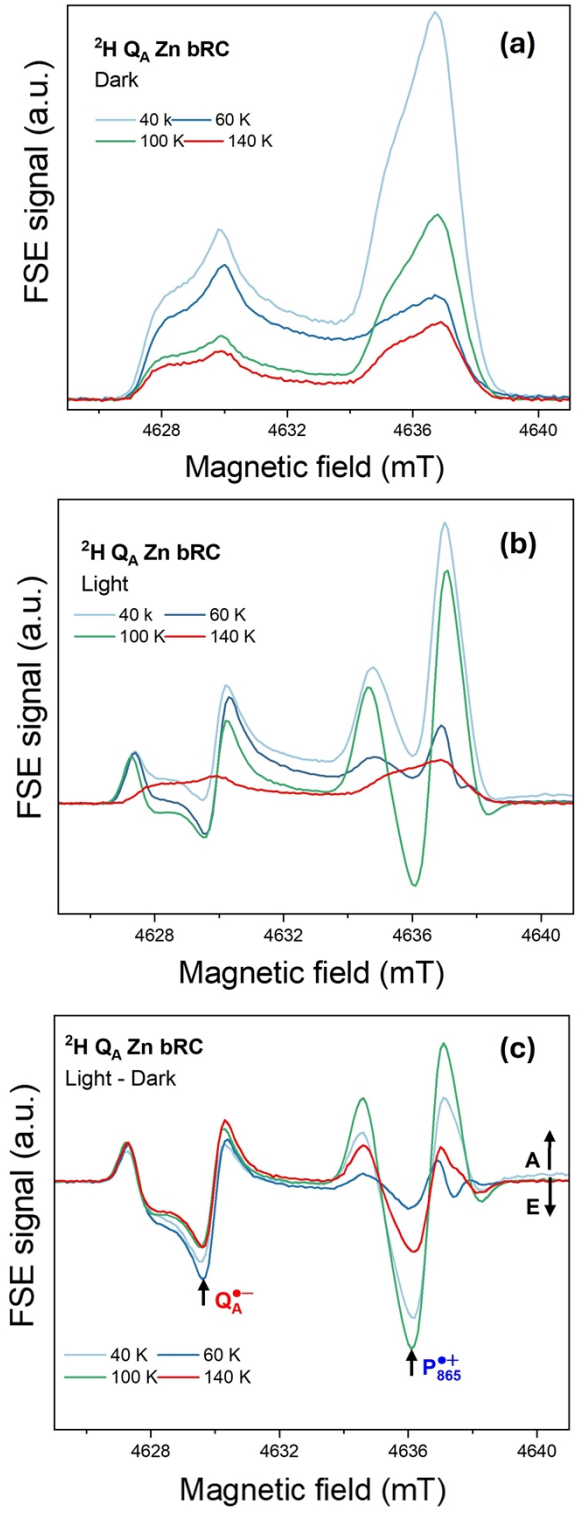

Figure S3: Temperature dependence of field-swept echo detected D-band (130 GHz) EPR spectra "light" (a), "dark" (b) and difference/light-dark (c) for Zn bRC with ^{2}H Q_A.

Two pulse ESEEM at D-band (130 GHz) were recorded for both Q_A^- and P_{865}^+ as indicated in Figure S1. Figure S4 shows dark, and difference (light-dark) obtained at different temperatures for ^{2}H Zn bRC. The echo decays were fit with a mono-exponential function. The dash lines in the figures correspond to these fits. Similar temperature dependence was also obtained for other samples (Figure S5 and S6).

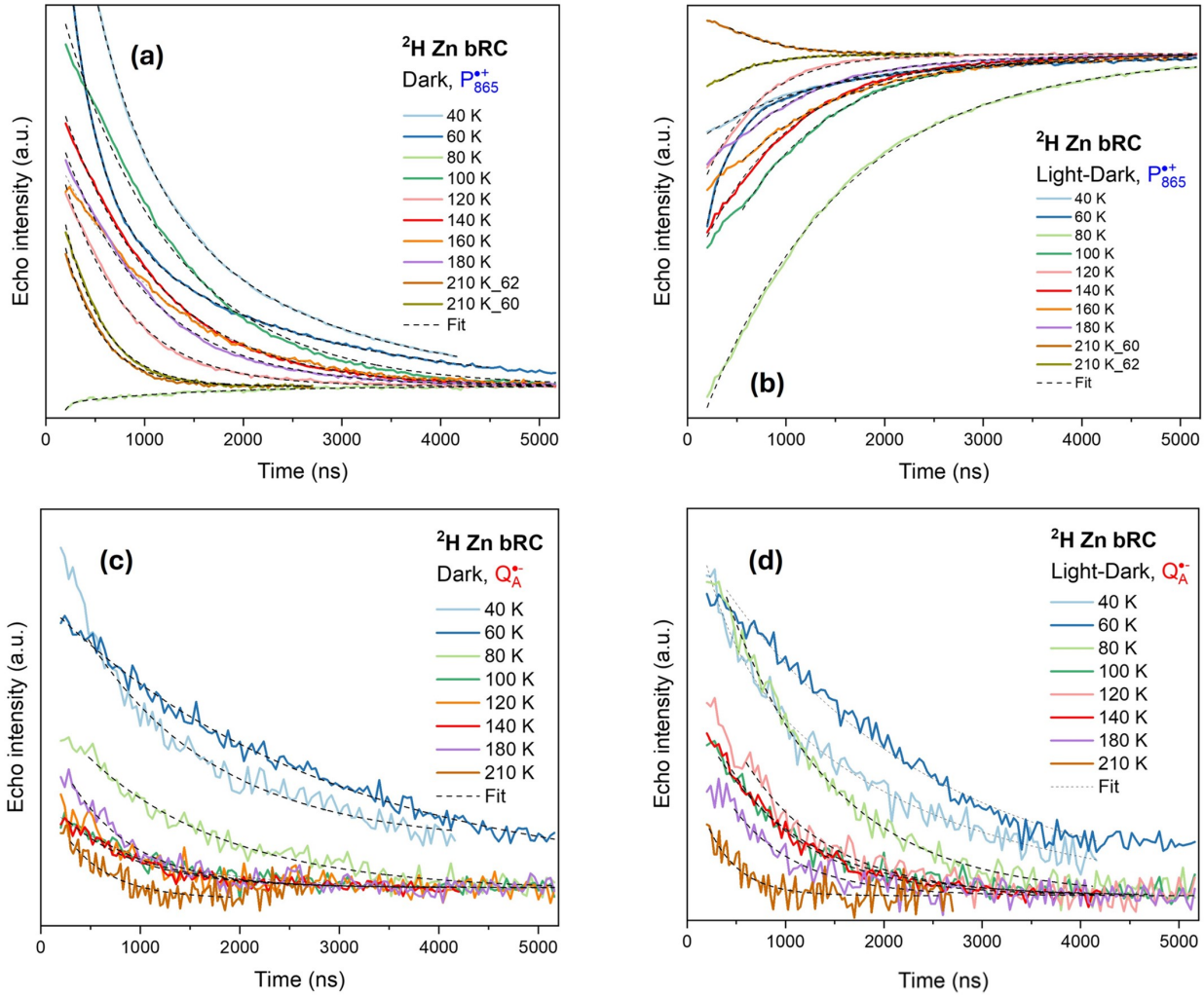

Figure S4: Experiment and mono-exponential fits of D band (130 GHz) 2-pulse ESEEM of ^{2}H Zn bRC. ESEEM plots here show experimental dark and difference (light-dark) for both Q_A^- and P_{865}^+. Time is 2τ.

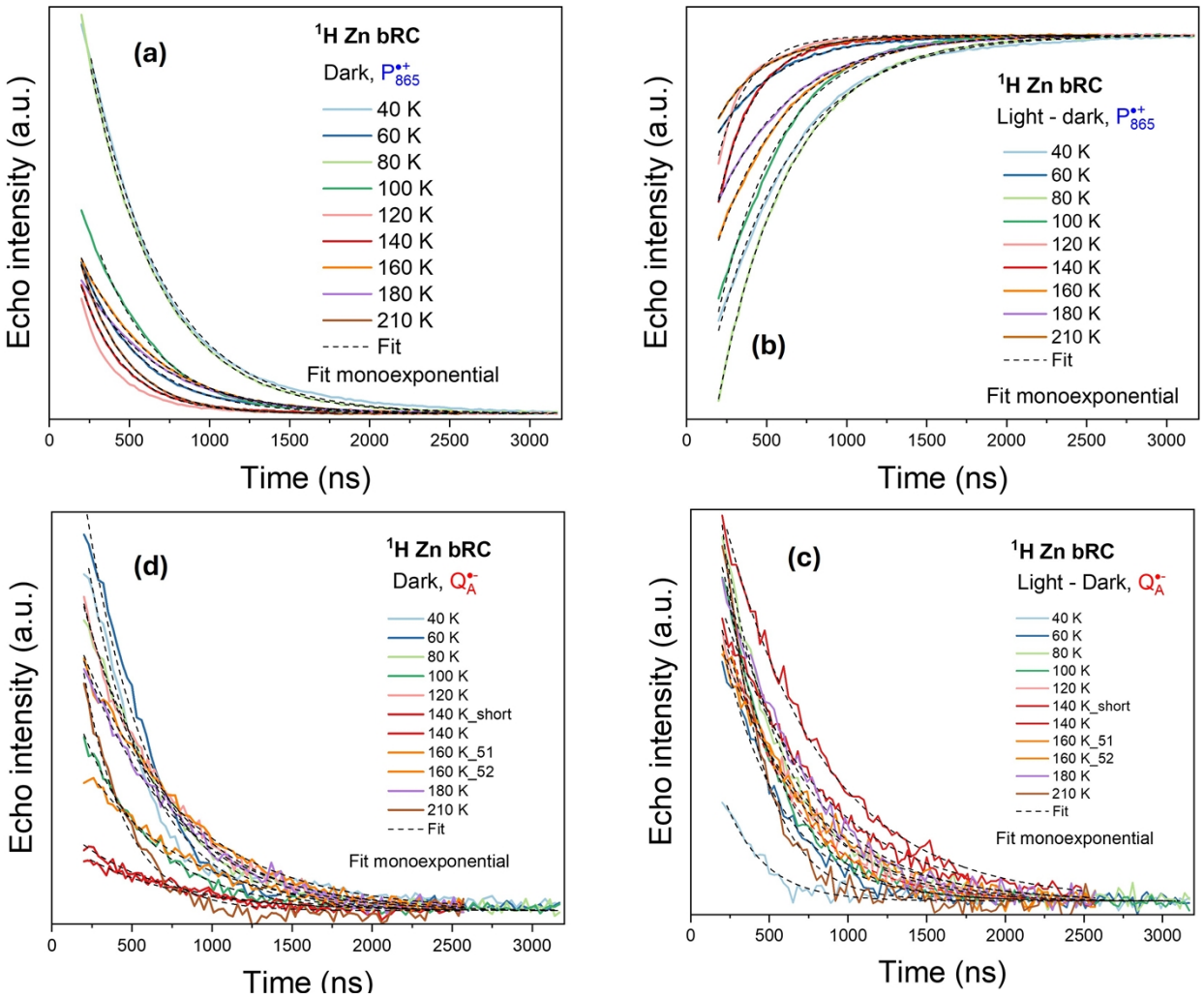

Figure S5: Experiment and mono-exponential fits of D band (130 GHz) 2-pulse ESEEM of ^{1}H Zn bRC. ESEEM plots here show experimental dark and difference (light-dark) for both Q_A^- and P^+_{865}. Time is 2τ.

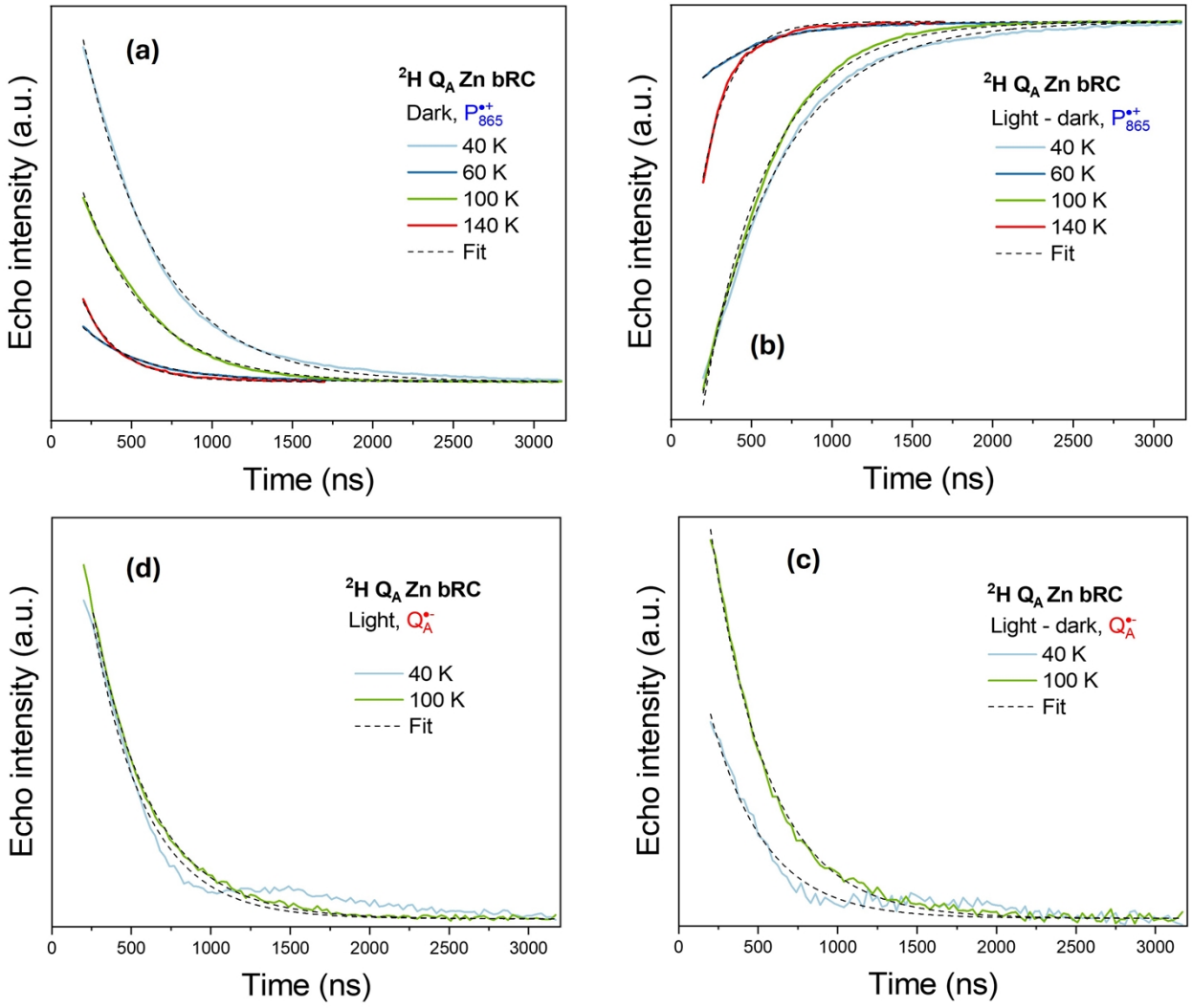

Figure S6: Experiment and mono-exponential fits of D band (130 GHz) 2-pulse ESEEM of Zn bRC with ^{2}H Q_A. ESEEM plots here show experimental dark and difference (light-dark) for both Q_A^- and P_{865}^+. Time is 2τ.

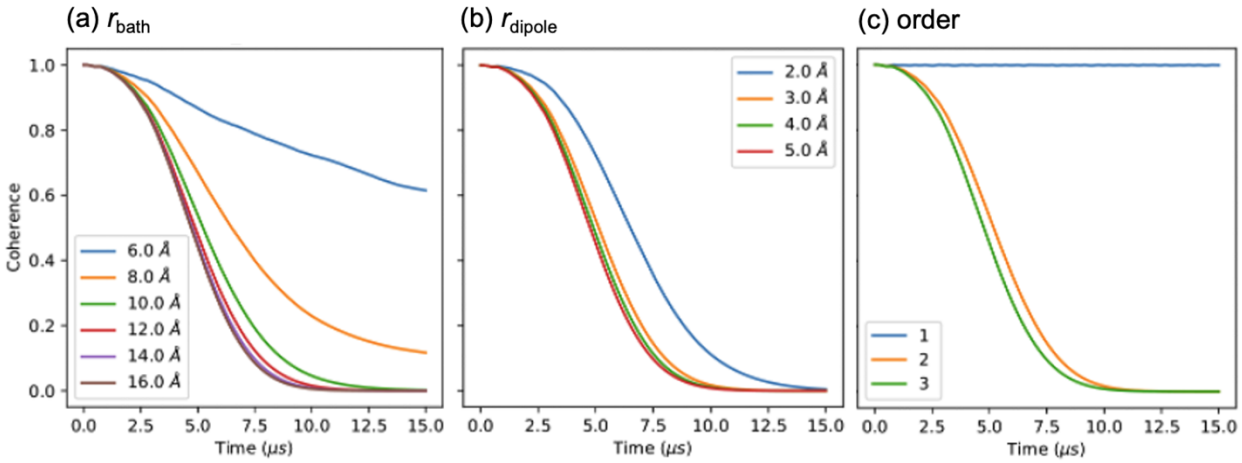

Figure S7: Sensitivity tests of key numerical parameters in the CCE calculations for the quinone site (U10) in the M chain of the photosynthetic reaction center of *R. sphaeroides*: (a) r_{bath}, the cutoff distance for the bath spin cluster measured from the electron spin position, (b) r_{dipole}, the cutoff distance for bath-spin–bath-spin interactions, and (c) the CCE order. When not varied, the parameters used are order = 3, $r_{bath} = 14$ Å, and $r_{dipole} = 5$ Å.

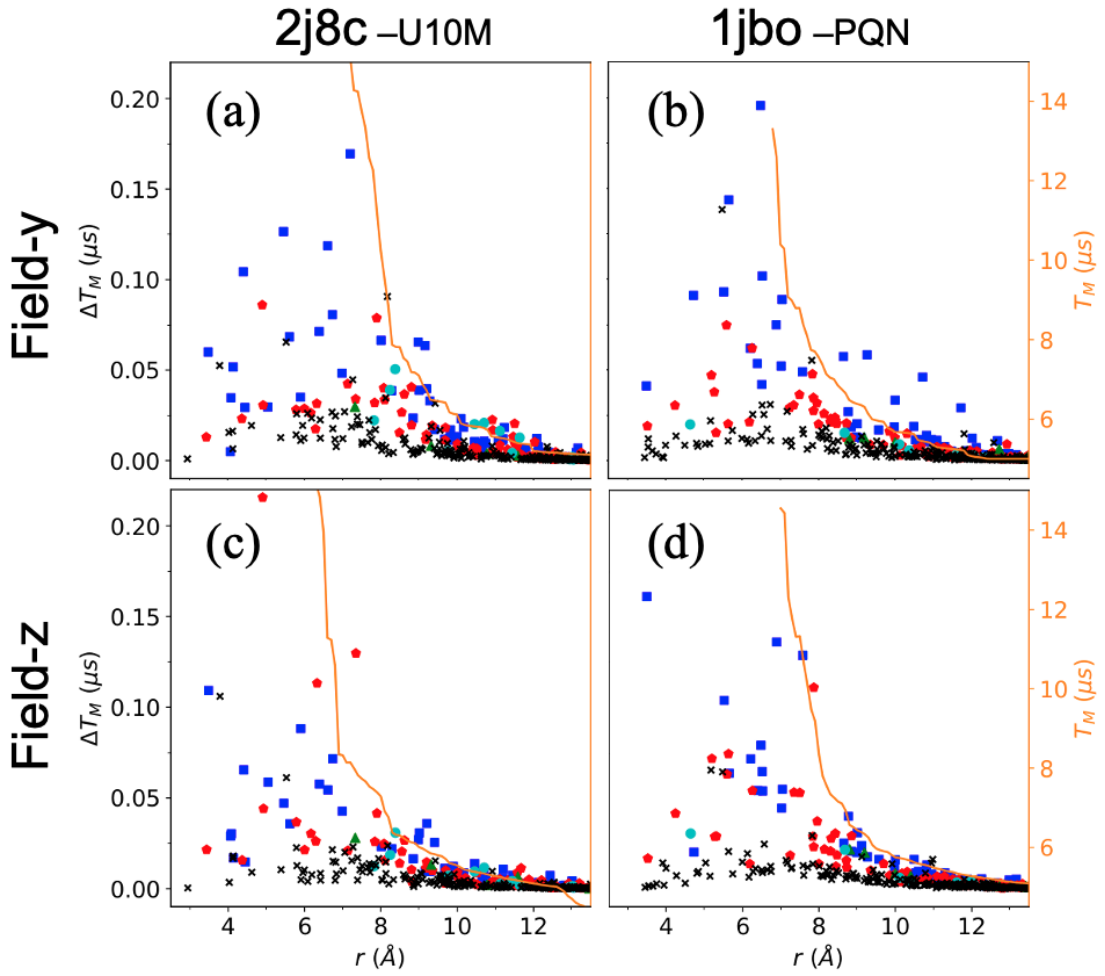

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Table S1. Calculated NSD coherence times T_M for the quinone electron spin for protonated and deuterated bRC. The unmodified results may be compared with the results of artificial calculations where the interactions of the protons or deuterons on the methyl groups were increased by factors of 10–100.

Methyl $^{1/2}$H interaction multiplier	T_M (μs)	
	Protonated	Deuterated
1 (unmodified)	5.61	68.9
10	1.29	23.7
20	0.798	11.1
30		7.47
40		6.11
50		5.30
100		3.64