Supporting Information to "Chirality Induced Spin Coherence in Electron Transfer Reactions"

Thomas P. Fay*

Department of Chemistry, University of Oxford, Physical and Theoretical Chemistry Laboratory, South Parks Road, Oxford, OX1 3QZ, UK

E-mail: tom.patrick.fay@gmail.com

1 The spin-orbit coupling interaction

In the main paper we simply state that the spin-orbit coupling term in a trunctated diabatic basis containing two charge transfer states is given by Eqs. (3) and (11). In the one electron case, the electronic state factorises into spatial and spin parts given by $|A, \sigma\rangle = \psi_A(\mathbf{r}|\mathbf{Q}) |\sigma\rangle$, where $\psi_A(\mathbf{r}|\mathbf{Q})$ is the nuclear configuration dependent diabatic state wavefunction. Importantly for bound electronic states these wavefunctions can always be chosen to be real-valued.¹ The full spin orbit coupling Hamiltonian for an *N* electron system is¹

$$\hat{H}_{\text{SOC}} = \sum_{i=1}^{N} \sum_{K} \xi_{K}(r_{iK}) \hat{\ell}_{iK} \cdot \hat{\mathbf{s}}_{i}, \qquad (S.1)$$

where $r_{iK} = |\mathbf{r}_i - \mathbf{Q}_K|$, $\xi_K(r_{iK})$ is a real-valued function of r_{iK} and $\hat{\ell}_{iK} = (\mathbf{r}_i - \mathbf{Q}_K) \times \hat{\mathbf{p}}_i$ is the orbital angular momentum of electron *i* about nucleus *K*, $\hat{\mathbf{s}}_i$ is the momentum operator for electron *i* and $\hat{\mathbf{s}}_i$ is the spin operator for electron *i*. The SOC matrix elements are then given by (noting that for one electron we can drop the electron labels *i*)

$$\langle A, \sigma | \hat{H}_{\text{SOC}} | A', \sigma' \rangle = \sum_{\alpha = x, y, z} \langle \sigma | \hat{s}_{\alpha} | \sigma' \rangle \int d\mathbf{r} \, \psi_j(\mathbf{r} | \mathbf{Q})^* \sum_K \xi_A(r_K) \hat{\ell}_{K, \alpha} \psi_{A'}(\mathbf{r} | \mathbf{Q}) \tag{S.2}$$

and therefore we identify the components of the spin orbit coupling vector $i \Lambda_{A,A'}(\mathbf{Q})$ as

$$i\Lambda_{A,A',\alpha}(\mathbf{Q}) = \int d\mathbf{r} \,\psi_A(\mathbf{r}|\mathbf{Q})^* \sum_K \xi_K(r_K) \hat{\ell}_{K,\alpha} \psi_{A'}(\mathbf{r}|\mathbf{Q}).$$
(S.3)

Noting that $\hat{\mathbf{p}} = -i\hbar \nabla_{\mathbf{r}}$, and that the diabatic wave functions are real-valued, this term must be zero for A = A' and for $A \neq A' \Lambda_{A,A',\alpha}(\mathbf{Q})$ is real valued with $\Lambda_{A,A',\alpha}(\mathbf{Q}) = -\Lambda_{A',A,\alpha}(\mathbf{Q})$. This immediately gives the diabatic basis spin-orbit coupling Hamiltonian given by Eq. (3) in the main text, when ψ_A is taken to be an orbital localised on D, ψ_D , and $\psi_{A'}$ is taken to be an orbital localised on A, ψ_A , and when we also invoke the Condon approximation i.e. assuming that these matrix elements have only a weak dependence on \mathbf{Q} for accessible conformations of the molecule.

This argument generalises to the many electron case as follows. For an arbitrary electronic state $|\Psi_A, S = 1/2, M_S\rangle$ with total spin quantum number S = 1/2 with real expansion coefficients in terms of Slater determinants of real-valued molecular orbitals, it is true that

$$\langle \Psi_A, 1/2, \pm 1/2 | \hat{H}_{\text{SOC}} | \Psi_B, 1/2, \pm 1/2 \rangle = i a_{A,B} \pm b_{A,B}$$
 (S.4)

$$\langle \Psi_A, 1/2, \pm 1/2 | \hat{H}_{\text{SOC}} | \Psi_B, 1/2, \pm 1/2 \rangle = \pm i c_{A,B}$$
 (S.5)

where $a_{A,B}$, $b_{A,B}$ and $c_{A,B}$ are real valued, and all 0 if A = B (see Ref. 2 for details). For two S = 1/2 diabatic states $|\Psi_0, 1/2, M_S\rangle = |D^{\bullet-}A, 1/2, M_S\rangle = |0, 1/2, M_S\rangle$ and $|\Psi_1, 1/2, M_S\rangle = |DA^{\bullet-}, 1/2, M_S\rangle = |1, 1/2, M_S\rangle$, the spin orbit coupling Hamiltonian in this truncated basis is therefore

$$\hat{V}_{\text{SOC}} = 2i \left(a_{0,1} \hat{S}_x + b_{0,1} \hat{S}_y + c_{0,1} \hat{S}_z \right) \left(|0\rangle \langle 1| - |1\rangle \langle 0| \right)$$
(S.6)

and we can therefore identify $\Lambda = -2(a_{0,1}, b_{0,1}, c_{0,1})$. These are implicitly dependent on **Q** but again we can invoke the Condon approximation to ignore this **Q** dependence.

In order to understand the two unpaired electron radical pair case, we will first consider a two electron system, in which orbitals $\psi_A(\mathbf{r}|\mathbf{Q})$ are occupied, and the total electronic states are denoted $|A, B, \sigma_1, \sigma_2\rangle$ which are the antisymmetrised states in which orbital *A* is occupied with spin σ_1 and orbital *B* is occupied with spin σ_2 ,

$$|A, B, \sigma_1, \sigma_2\rangle = \frac{1}{\sqrt{2}} \bigg(\psi_A(\mathbf{r}_1) \psi_B(\mathbf{r}_2) |\sigma_1\rangle_1 |\sigma_2\rangle_2 - \psi_B(\mathbf{r}_1) \psi_A(\mathbf{r}_2) |\sigma_2\rangle_1 |\sigma_1\rangle_2 \bigg), \qquad (S.7)$$

where $|\sigma\rangle_j$ is a spin state of electron *j*, and **r**_j refer to the coordinates of electron *j*, and we have dropped the explicit dependence on **Q** for brevity. In our case we consider the coupling between a state $|0\rangle$ where one electron is the donor HOMO ψ_D , and one in the donor LUMO ψ_{D^*} (which corresponds to the photoexcited precursor state), and a state $|1\rangle$ where one electron is in the donor HOMO ψ_D and one in the acceptor LUMO ψ_A (which corresponds to the charge separated radical pair state). Because the spin orbit coupling matrix element between identical orbitals is zero, the only non-zero SOC matrix elements between these states can be written as

$$\langle \mathbf{D}^{*}, \mathbf{D}, \sigma_{1}, \sigma_{2} | \hat{H}_{\text{soc}} | \mathbf{A}, \mathbf{D}, \sigma_{1}', \sigma_{2}' \rangle = \sum_{K, \alpha} \langle \sigma_{2} | \sigma_{2}' \rangle$$

$$\times \langle \sigma_{1} | \hat{s}_{\alpha} | \sigma_{1}' \rangle \int d\mathbf{r} \psi_{\mathbf{D}^{*}}(\mathbf{r})^{*} \xi_{K}(r_{K}) \hat{\ell}_{K, \alpha} \psi_{\mathbf{A}}(\mathbf{r}).$$

$$(S.8)$$

As before the spin-orbit coupling integral between ψ_{D^*} and ψ_A is purely imaginary valued. We define the effective electron spin operator $\hat{\mathbf{S}}_1$ on the truncated set of states $|A, B, \sigma_1, \sigma_2\rangle$ in terms of the matrix elements of an S = 1/2 spin operator $\hat{\mathbf{s}}$ i.e. $\langle A, B, \sigma_1, \sigma_2 | \hat{\mathbf{S}}_1 | A', B', \sigma'_1, \sigma'_2 \rangle = \langle \sigma_1 | \hat{\mathbf{s}} | \sigma'_1 \rangle \delta_{A,A'} \delta_{B,B'} \delta_{\sigma_2,\sigma'_2}$. We can now construct the spin orbit coupling operator on the truncated set of states $|0\rangle = |0, \mathbf{S}\rangle = \frac{1}{\sqrt{2}} (|\mathbf{D}^*, \mathbf{D}, \alpha, \beta\rangle - |\mathbf{D}^*, \mathbf{D}, \beta, \alpha\rangle)$ and $|1, \sigma_1, \sigma_2\rangle = |A, \mathbf{D}, \sigma_1, \sigma_2\rangle$, exactly as is given in Eq. (11) in the main text.

Again we can generalise this to a many electron system by noting that for arbitrary electronic states $|\Psi_A, S, M_S\rangle$ which have total electronic spin quantum number S = 0 or S = 1 and total electron spin projection quantum number M_S , provided they can be expanded as a real linear combination of Slater determinants of real valued molecular orbitals, then the spin orbit coupling matrix elements have the following properties²

$$\langle \Psi_A, 0, 0 | \hat{H}_{\text{soc}} | \Psi_B, 1, \pm 1 \rangle = \mp i a_{A,B} + b_{A,B}$$
(S.9)

$$\langle \Psi_A, 0, 0 | \hat{H}_{\text{soc}} | \Psi_B, 1, 0 \rangle = i c_{A,B}, \qquad (S.10)$$

where again the constants are all real-valued. Now considering just a subset of diabatic states $|\Psi_0, 0, 0\rangle = |D^*A, 0, 0\rangle = |0, 0, 0\rangle$, and $|\Psi_1, S, M_S\rangle = |D^{\bullet+}A^{\bullet-}, S, M_S\rangle = |1, S, M_S\rangle$ which correspond to the precursor state and the set of S = 0 and S = 1 radical pair states, and ignoring the spin-orbit coupling between radical pair states (which vanishes for large radical separations), the spin orbit coupling operator can be written as in Eq. (11) of the main text with the above definition

of the effective electron spin operator $\hat{\mathbf{S}}_1$ with

$$\mathbf{\Lambda} = (2\sqrt{2}a_{0,1}, 2\sqrt{2}b_{0,1}, 2c_{0,1}). \tag{S.11}$$

2 Second order master equation theory

In this appendix we briefly summarise the master equation used to obtain the spin density operator master equations in the main text. Further details can be found in Ref. 3. Our starting point is the Nakajima-Zwanzig equation^{4,5} for a projected density operator $\mathcal{P}\hat{\rho}(t)$, which contains the spin density operators,³

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathcal{P}\hat{\rho}(t) = \mathcal{P}\mathcal{L}\mathcal{P}\hat{\rho}(t) + \int_0^t \mathcal{K}(t-\tau)\mathcal{P}\hat{\rho}(\tau)\,\mathrm{d}\tau\,,\tag{S.12}$$

where $\mathcal{L} = -(i/\hbar)[\hat{H}, \cdot]$ is the total Liouvillian for the system and $\mathcal{K}(t)$ is a memory kernel which contains the effects of parts of the system removed by projecting the density operator with \mathcal{P} . We assume that the charge transfer coupling is weak, and thus the initial state of the system can be assumed to a be a local equilibrium state on each diabatic surface, $\hat{\rho}(0) = \sum_{j} \hat{\Pi}_{j} \hat{\sigma}_{j}(0) \hat{\rho}_{jn}$, with $\hat{\rho}_{jn} = e^{-\beta \hat{H}_{jn}} / \text{Tr}_{n} [e^{-\beta \hat{H}_{jn}}]$ and $\hat{\sigma}_{j}(0)$ being the initial spin density operator for state *j*. Analogous to previous work, we define $\mathcal{P} = \sum_{j} \hat{\rho}_{jn} \text{Tr}_{n} [\hat{\Pi}_{j} \cdot \hat{\Pi}_{j}]$, such that $\mathcal{P}\hat{\rho}(0) = \hat{\rho}(0)$ and we expand the kernel $\mathcal{K}(t)$ to second order in the coupling Γ .

The approximate treatment of the spin-orbit and diabatic coupling is based on approximating the kernel in the Nakajima-Zwanzig equation, which is given by

$$\mathcal{K}(t) = \mathcal{P}\mathcal{L}e^{(1-\mathcal{P})\mathcal{L}t}(1-\mathcal{P})\mathcal{L}\mathcal{P}.$$
(S.13)

Expanding this up to second order in the spin-orbit coupling and diabatic coupling, and ignoring

the spin terms $\hat{H}_{j\mathrm{s}}$ in the kernel gives the following approximation to $\mathcal{K}(t)$

$$\mathcal{K}(t)\mathcal{P}\hat{\rho} \approx -\frac{1}{\hbar^2} \sum_{j} \hat{\rho}_{jn} \operatorname{Tr}_{n} \left[\hat{\Pi}_{j} [\hat{V}, [\hat{V}^{n}(t), \mathcal{P}\hat{\rho}]] \hat{\Pi}_{j} \right]$$
(S.14)

where $\hat{V} = \hat{V}_{DC} + \hat{V}_{SOC}$ and $\hat{V}^n(t) = e^{-i\hat{H}_n t/\hbar} \hat{V} e^{+i\hat{H}_n t/\hbar}$, where $\hat{H}_n = \hat{H}_{0n}\hat{\Pi}_0 + \hat{H}_{1n}\hat{\Pi}_1$. We also invoke the Markovian approximation to the time-convolution term, in which we assume the decay time of the kernel is much faster than the characteristic timescales of the spin and population dynamics, and therefore we can approximate the time convolution in the Nakajima-Zwanzig equation as⁶

$$\int_0^t \mathcal{K}(t-\tau)\mathcal{P}\hat{\rho}(\tau)\,\mathrm{d}\tau \approx \int_0^\infty \mathcal{K}(\tau)\,\mathrm{d}\tau\,\mathcal{P}\hat{\rho}(t). \tag{S.15}$$

Using this approximation we recover the incoherent kinetic description of the electron transfer processes.

By using these approximations and expanding the double commutator in the second order kernel, one arrives at the master equations given above, with $k_{\rm f}$ given by the standard Fermi's Golden rule rate expression,⁷

$$k_{\rm f} = \frac{2\Gamma^2}{\hbar^2} \operatorname{Re} \int_0^\infty \operatorname{Tr}_{\rm n} [e^{+i\hat{H}_{0\rm n}t/\hbar} e^{-i\hat{H}_{1\rm n}t/\hbar} \hat{\rho}_{0\rm n}] \,\mathrm{d}t$$
(S.16)

and similarly $k_{\rm b}$ is given by

$$k_{\rm b} = \frac{2\Gamma^2}{\hbar^2} \operatorname{Re} \int_0^\infty \operatorname{Tr}_{\rm n} \left[e^{+i\hat{H}_{\rm ln}t/\hbar} e^{-i\hat{H}_{\rm 0n}t/\hbar} \hat{\rho}_{\rm 1n} \right] \mathrm{d}t \tag{S.17}$$

and the δJ term in the radical pair master equation is

$$\delta J = \frac{\Gamma^2}{2\hbar} \operatorname{Im} \int_0^\infty \operatorname{Tr}_n[e^{+i\hat{H}_{0n}t/\hbar} e^{-i\hat{H}_{1n}t/\hbar} \hat{\rho}_{0n}] \,\mathrm{d}t \,.$$
(S.18)

The derivation of these expressions is described in detail in Ref. 3. We note that going to higher orders in $\hat{\Gamma}$, the S = 1/2 system master equation is unchanged, but at fourth order and above in Γ in

the radical pair master equation a decoherence term of the following form will also appear in the master equation,

$$k_{\rm D} \left(\hat{U} \hat{P}_{\rm S} \hat{U}^{\dagger} \hat{\sigma}_{1\rm s}(t) \hat{U} \hat{P}_{\rm S} \hat{U}^{\dagger} - \frac{1}{2} \left\{ \hat{U} \hat{P}_{\rm S} \hat{U}^{\dagger}, \hat{\sigma}_{1\rm s}(t) \right\} \right). \tag{S.19}$$

Also the expressions for the master equation parameters k_f , k_b and δJ must be corrected at higher orders in Γ , but the general form of the master equation remains unchanged other than the additional decoherence term. In this treatment the coupling of electronic and vibrational degrees of freedom is treated exactly, and the only approximations invoked are the perturbative treatment of electronic state coupling, the neglect of spin interactions on the electronic state transition kernel, and the Markovian approximation.

We note finally that in all of the above, we have used the Condon approximation, in which we assume Δ and Λ are independent of \mathbf{Q} . However in reality these terms will be nuclear configuration dependent and in general they will have a different dependence on \mathbf{Q} . Accounting for this will introduce terms which lead to spin decoherence on electron transport, i.e. terms which only transfer populations and not spin coherences.

3 OOP-ESEEM Signals for chiral radical pairs

Here we outline the derivation of the OOP-ESEEM signal for a chiral radical pair formed by SOCT. We consider the final time dependent *x* channel FID signals in the rotating frame $f_x(t) =$ $\text{Tr}_s[\hat{S}_x\hat{\sigma}_s(t)]$. Here $\sigma_s(t)$ is the radical pair spin density operator (denoted $\hat{\sigma}_{1s}(t)$ above) after the OOP-ESEEM pulse sequence, and $\hat{S}_{\alpha} = \hat{S}_{1\alpha} + \hat{S}_{2\alpha}$. For simplicity in finding $f_x(t)$, we will invoke the high field (secular) approximation for the radical pair spin Hamiltonian in the rotating frame,⁸

$$\frac{\hat{H}_{s}}{\hbar} = \Omega_{1}\hat{S}_{1,z} + \Omega_{2}\hat{S}_{1,z} - 2J\hat{\mathbf{S}}_{1}\cdot\hat{\mathbf{S}}_{2} + \frac{1}{2}d\left(\hat{S}_{z}^{2} - \frac{1}{3}\hat{S}^{2}\right) + \sum_{i=1}^{2}\sum_{k=1}^{N_{i}}a_{i,k}\hat{\mathbf{I}}_{i,k}\cdot\hat{\mathbf{S}}_{i}.$$
(S.20)

From right to left these terms describe the Zeeman interactions of two electron spins (relative to the rotating frame frequency), the scalar electron spin coupling, the dipolar coupling (with $d = D(3\cos^2(\xi) - 1)$, where *D* is the dipolar coupling constant), and the hyperfine coupling terms.

For simplicity we ignore chiral spin-orbit effects on evolution of the radical pair spin density, for example terms of the form $2\delta J \hat{U} \hat{P}_{\rm S} \hat{U}^{\dagger}$, and as a further simplification we will ignore any decay processes of the radical pair. This means we will assume the radical pair spin density operator evolves under the equation,

$$\frac{\mathrm{d}}{\mathrm{d}t}\hat{\sigma}_{\mathrm{s}}(t) = -\frac{i}{\hbar}\left[\hat{H}_{\mathrm{s}},\hat{\sigma}_{\mathrm{s}}(t)\right] = \mathcal{L}_{\mathrm{s}}\hat{\sigma}_{\mathrm{s}}(t). \tag{S.21}$$

The action of a perfect (instantaneous) microwave pulse on the spin density operator by an angle φ , about an axis α is $\mathcal{U}_{\alpha}(\varphi)\hat{\sigma}_{s} = e^{-i\varphi\hat{S}_{\alpha}}\hat{\sigma}_{s}e^{+i\varphi\hat{S}_{\alpha}}$. Finally we assume the nuclear spins are in the infinite temperature thermal equilibrium state, i.e. a completely mixed state, at t = 0, therefore the initial spin density operator of a chiral radical pair is $\hat{\sigma}_{s}(0) = (1/Z)\hat{U}\hat{P}_{s}\hat{U}^{\dagger}$, where Z is the dimensionality of the nuclear spin Hilbert space. The spin density operator in the rotating frame at a time t of the echo, $\hat{\sigma}_{s}(t)$, is then found as

$$\hat{\sigma}_{s}(t) = e^{\mathcal{L}_{s}t} e^{\mathcal{L}_{s}\tau} \mathcal{U}_{x}(\pi) e^{\mathcal{L}_{s}\tau} \mathcal{U}_{x}(\pi/4) \hat{\sigma}_{s}(0).$$
(S.22)

We will now use this to calculate the OOP-ESEEM signal when the radical pair is oriented such that **n** is parallel to the lab *z* axis. We note that the Hamiltonian commutes with $\hat{I}_{i,k,z}$, and therefore its eigenstates can be written as $|n, \mathbf{M}\rangle$, where **M** denotes the set of *z* projection quantum numbers for the nuclear spins. For simplicity, we make the weak-coupling approximation here, $|\Omega_1 - \Omega_2| \gg |J - d/6|$, and then using standard expressions for the radical pair eigenstates,⁸ one finds the total x channel OOP-ESEEM signal to be

$$f_{x}(t) = \frac{\sin(\theta)}{Z} \sum_{\mathbf{M}} \cos(\bar{\omega}_{\mathbf{M}}t) \sin\left(\left(\frac{d}{3} - J\right)(t + 2\tau)\right)$$

$$\times \left(\sqrt{2}\cos(\theta)\sin(\Delta\omega_{\mathbf{M}}t) - \sin(\theta)\cos(\Delta\omega_{\mathbf{M}}t)\right)$$
(S.23)

where $\bar{\omega}_{\mathbf{M}} = (\omega_{1,\mathbf{M}} + \omega_{2,\mathbf{M}})/2$, $\Delta \omega_{\mathbf{M}} = (\omega_{1,\mathbf{M}} - \omega_{2,\mathbf{M}})/2$ $\omega_{i,\mathbf{M}} = \Omega_i + \sum_{k=1}^{N_i} a_{i,k} M_{i,k}$. In the semiclassical limit, where the number of hyperfine coupled nuclear spins is larger, we can replace $\sum_{k=1}^{N_i} a_{i,k} M_{i,k} \rightarrow h_{i,z}$, and $\frac{1}{Z} \sum_{\mathbf{M}} \rightarrow \prod_{i=1}^2 \int_{-\infty}^{\infty} dh_{i,z} P_{i,z}(h_{i,z})$, where the probability distribution is given by⁹

$$P_{i,z}(h_{i,z}) = \frac{\tau_i}{\sqrt{2\pi}} \exp\left(-\frac{1}{2} \left(h_{i,z}\tau_i\right)^2\right)$$
(S.24)

where $\tau_i^{-2} = \frac{1}{3} \sum_{k=1}^{N_i} a_{i,k}^2 I_{i,k} (I_{i,k} + 1)$. This gives the expression for $f_x(t)$ in the weak coupling limit given in the main text.

Beyond the weak coupling limit, but still within the high field approximation, we can obtain analytical expressions for the *x* channel FID signal,

$$f_{x}(t) = \frac{1}{Z} \sum_{\mathbf{M}} \frac{1}{2} \cos(t\bar{\omega}_{\mathbf{M}}) \left(2\sin^{2}(\theta) \cos(\Delta_{\mathbf{M}}\tau) \sin((J - d/3)(t + 2\tau)) \cos(\Delta_{\mathbf{M}}(t + \tau)) - 2\sin^{2}(\theta) \cos(4\phi_{\mathbf{M}}) \sin(\Delta_{\mathbf{M}}\tau) \sin((J - d/3)(t + 2\tau)) \sin(\Delta_{\mathbf{M}}(t + \tau)) - 2\sin^{2}(\theta) \cos(2\phi_{\mathbf{M}}) \cos((J - d/3)(t + 2\tau)) \sin(\Delta_{\mathbf{M}}(t + 2\tau)) - \sqrt{2}\sin(\theta) \left(\sin(2\phi_{\mathbf{M}}) \sin(\Delta_{\mathbf{M}}t) \sin((J - d/3)(t + 2\tau)) + \sin(4\phi_{\mathbf{M}}) \sin(\Delta_{\mathbf{M}}\tau) \cos((J - d/3)(t + 2\tau)) \sin(\Delta_{\mathbf{M}}(t + \tau))) \right)$$
(S.25)

where $\bar{\omega}_{\mathbf{M}} = (\omega_{1,\mathbf{M}} + \omega_{2,\mathbf{M}})/2$, $\Delta_{\mathbf{M}} = \sqrt{\Delta \omega_{\mathbf{M}}^2 + (J + d/6)^2}$, $\tan(2\phi_{\mathbf{M}}) = \Delta \omega_{\mathbf{M}}/(J + d/6)$, $\Delta \omega_{\mathbf{M}} = (\omega_{1,\mathbf{M}} - \omega_{2,\mathbf{M}})/2$, and $\omega_{i,\mathbf{M}} = \Omega_i + \sum_{k=1}^{N_i} a_{i,k} M_{i,k}$. By making the same semiclassical replacement as above, we can numerically evaluate the semiclassical FID signals as a function of *t*. We can then numerically integrate these for a range of τ values to yield $F_x(\tau)$.

In the weak coupling, semiclassical limit the integrated FID signal can be found analytically as a function of τ by integrating Eq. (18) as $F_x(\tau) = \int_0^\infty f_x(t) dt$,

$$\begin{split} F_{x}(\tau) &= \frac{\sqrt{\pi}}{8} \Biggl[\tau_{1} \left(e^{-\tau_{1}^{2} (J-d/3+\Omega_{1})^{2}/2} - e^{-\tau_{1}^{2} (J-d/3-\Omega_{1})^{2}/2} \right) \Biggr] \cos(2(J-d/3)\tau) \sin(\theta) \\ &+ \tau_{2} \left(e^{-\tau_{2}^{2} (J-d/3-\Omega_{2})^{2}/2} - e^{-\tau_{2}^{2} (J-d/3+\Omega_{2})^{2}/2} \right) \Biggr] \cos(2(J-d/3)\tau) \sin(\theta) \\ &+ \frac{1}{4} \Biggl[\tau_{1} \left(D_{+} (\tau_{1} (J-d/3-\Omega_{1})/\sqrt{2}) - D_{+} (\tau_{1} (J-d/3+\Omega_{1})/\sqrt{2}) \right) \Biggr] \\ &+ \tau_{2} \left(D_{+} (\tau_{2} (J-d/3+\Omega_{2})/\sqrt{2}) - D_{+} (\tau_{2} (J-d/3-\Omega_{2})/\sqrt{2}) \right) \Biggr] \sin(2(J-d/3)\tau) \sin(\theta) \\ &+ \frac{1}{4} \sqrt{\frac{\pi}{2}} \Biggl[\tau_{1} \left(e^{-\tau_{1}^{2} (J-d/3+\Omega_{1})^{2}/2} + e^{-\tau_{1}^{2} (J-d/3-\Omega_{1})^{2}/2} \right) \Biggr] \\ &+ \tau_{2} \left(e^{-\tau_{2}^{2} (J-d/3-\Omega_{2})^{2}/2} + e^{-\tau_{2}^{2} (J-d/3+\Omega_{2})^{2}/2} \right) \Biggr] \sin(2(J-d/3)\tau) \sin^{2}(\theta) \\ &+ \frac{1}{2\sqrt{2}} \Biggl[\tau_{1} \left(D_{+} (\tau_{1} (J-d/3-\Omega_{1})/\sqrt{2}) + D_{+} (\tau_{1} (J-d/3+\Omega_{1})/\sqrt{2}) \right) \Biggr] \\ &+ \tau_{2} \left(D_{+} (\tau_{2} (J-d/3+\Omega_{2})/\sqrt{2}) + D_{+} (\tau_{2} (J-d/3-\Omega_{2})/\sqrt{2}) \right) \Biggr] \cos(2(J-d/3)\tau) \sin^{2}(\theta) \end{aligned}$$
(S.26)

where $D_+(x) = e^{-x^2} \int_0^x e^{\lambda^2} d\lambda$ is the Dawson function.

4 Magnetic field effects

Chirality induced spin coherence in oriented radical pairs could also manifest in magnetic field effect experiments, in which quantum yields of spin selective radical pair reactions, or the lifetime of radical pair state, are measured as the applied magnetic field strength is varied, for example by transient absorption spectroscopy.^{10,11}

As a simple illustrative example, as above, we consider a radical pair oriented such that the spin orbit coupled charge transport vector $\mathbf{\Lambda}$ is aligned parallel to the applied magnetic field. We also take the high field limit where the Δg mechanism dominates the singlet-triplet interconversion. Treating the spin selective recombination process with the standard Haberkorn reaction term in the

master equation for $\hat{\sigma}_{s}(t)$, i.e. $-\{(k_{s}/2)\hat{P}_{s} + (k_{T}/2)\hat{P}_{T}, \hat{\sigma}_{s}(t)\}$, and assuming that $|\Omega_{1} - \Omega_{2}| \gg |J|, |D|, |k_{s} - k_{T}|, 1/\tau_{i}$, we can use Eq. (S.20), to find the triplet quantum yield for a chiral radical pair, $\Phi_{T} = k_{T} \int_{0}^{\infty} \text{Tr}_{s}[\hat{P}_{T}\hat{\sigma}_{s}(t)] dt$ in the high field limit,

$$\Phi_{\rm T} = k_{\rm T} \frac{4\Delta\Omega^2 - \bar{k}^2 \cos(2\theta) + \bar{k}^2 - 2\Delta\Omega\bar{k}\sin(2\theta)}{2\bar{k}\left(\bar{k}^2 + 4\Delta\Omega^2\right)},$$
(S.27)

where $\Delta\Omega = \Omega_1 - \Omega_2$ and $\bar{k} = (k_{\rm S} + k_{\rm T})/2$. The term proportional to $\sin(2\theta)$ leads to a chirality dependent triplet quantum yield, and from this the difference in the triplet yields between two enantiomers is maximised when $|\Delta\Omega| = \bar{k}/2$.

Of course, the quantitative interpretation of any real experiment would require more detailed quantum mechanical modelling, accounting fully for hyperfine interactions and spin relaxation, ^{12–14} but this simple result demonstrates that chirality induced spin coherence could in principle lead to chirality dependent magnetic field effects on the reactions of oriented radical pairs.

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