

Interradical motion can push magnetosensing precision toward quantum limits

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Magnetosensitive spin-correlated radical pairs offer a promising platform for noise-robust quantum metrology; however, unavoidable interrational interactions, such as electron-electron dipolar and exchange couplings, alongside deleterious perturbations resulting from intrinsic radical motion, typically degrade their potential as magnetometers. In contrast, we show how structured molecular motion modulating interrational interactions in a *live* chemical sensor in cryptochrome can, in fact, increase sensitivity and, moreover, push precision in estimating magnetic field directions closer to the quantum Cramér-Rao bound. The latter is evident both on average over the magnetoreceptor array and, to an even greater extent, for the subset of best-performing magnetoreceptors, thus suggesting near-optimal metrological performance. Remarkably, this approach to optimality is amplified under environmental noise and persists with increasing complexity of the spin system, suggesting that perturbations inherent to such natural systems have enabled them to operate closer to the quantum limit to more effectively extract information from the weak geomagnetic field. This insight opens the possibility of channeling the underlying physical principles of motion-induced modulation of electron spin-spin interactions toward devising efficient handles over emerging molecular quantum information technologies.

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I. INTRODUCTION

The sensitivity of spin-correlated radical pairs (SCRPs) to weak magnetic fields presents exciting avenues for potential applications across quantum technologies and biophysics [1–7]. In a physiological context, SCRPs in the protein cryptochrome are widely considered to be candidate magnetoreceptors of a chemical compass sense for migratory navigation [8–11]. Such SCRPs function in noisy and complex condensed-phase regimes at ambient temperature while subjected to strong interrational interactions, such as electron-electron dipolar (EED) and exchange couplings, and environmentally induced structural fluctuations [12–17]. In engineered biradicals [18–21], such effects usually suppress the coherent singlet-triplet interconversion that underpins their metrological sensitivity. This raises fundamental questions: How close to quantum limits could SCRPs operate under noisy biological conditions? And could nature have optimized

SCRPs in the open [22–25] to enable optimal magnetosensitivity?

Recent work comparing SCRPs models in terms of the quantum Cramér-Rao bound (QCRB) [26], which sets a fundamental limit on parameter-estimation precision [27–29], showed a partial approach toward the QCRB in the limits of quantum system complexity, i.e., when increasing the number of hyperfine couplings toward biologically representative numbers; however, the precision fell short of that of the QCRB by more than an order of magnitude. In this work, we demonstrate that intrinsic interrational motion can drive magnetosensing in SCRPs to over 90% of the quantum limit, achieving up to sub-degree angular precision, challenging the prevalent notion that environmental noise and molecular motion would preclude sensitivity near the quantum limit. Specifically, we consider driven radical-pair systems, motivated by prior findings showing how Landau-Zener-Stückelberg-Majorana transitions [30] can enhance chemical-signal contrast [31], with similarly periodic driving also having been shown to boost precision in quantum thermometry [32]. We establish quantum limits by considering the relationship between radical motion and the QCRB for the flavin-adenine dinucleotide/tryptophan radical pair ($\text{FAD}^{\bullet-}/\text{W}_C^{*+}$) as a representative radical pair in cryptochrome, with our results informing more broadly about

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systems subject to electron spin-spin interactions, such as EED and/or exchange couplings.

II. DRIVEN RADICAL-PAIR MODEL

We consider radical pairs interacting with an external magnetic field and magnetic nuclei via Zeeman and hyperfine interactions, respectively, as shown in Figs. 1(a)–1(c). The time-independent part of the Hamiltonian defining these interactions is

$$\begin{aligned}\hat{H}_0 &= \hat{H}_{\text{Zeeman}} + \hat{H}_{\text{Hyperfine}} \\ &= \sum_i \vec{\omega}_i \cdot \hat{\mathbf{S}}_i + \sum_i \sum_j \hat{\mathbf{S}}_i \cdot \mathbf{A}_{ij} \cdot \hat{\mathbf{I}}_{ij},\end{aligned}\quad (1)$$

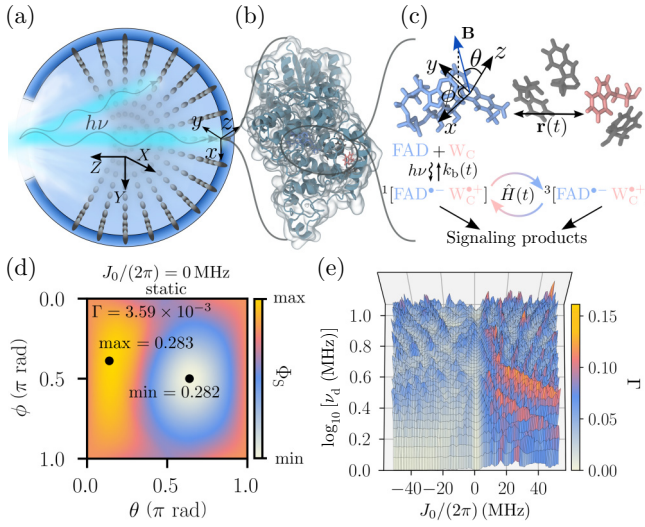


FIG. 1. Magnetosensing modulated by interrational motion. (a) Schematic of a magnetosensitive array [in the eye frame (X, Y, Z)], composed of magnetoreceptor units [gray spheroids; molecular frame (x, y, z)] distributed with fixed orientations along the retina. Incident blue light photoexcites radical pairs within the magnetoreceptors. (b) Blue-light-activated magnetoreceptor protein cryptochrome. (c) Within cryptochrome, an $\text{FAD}^{\bullet-}/\text{W}_C^+$ radical pair is created in the singlet state ($^1[\cdot]$) and interconverts with triplet states ($^3[\cdot]$) via the interrational-separation-modulated Hamiltonian $\hat{H}(r(t))$, which includes Zeeman, hyperfine, exchange, and electron-electron dipolar (EED) interactions. Spin-selective recombination occurs with rate $k_b(t)$, while signaling products form at a rate k_f . The Zeeman interaction imparts magnetosensitivity depending on the field orientation (θ, ϕ) in the molecular frame. (d) Angular dependence of the singlet yield Φ_S and relative anisotropy $\Gamma = [\max(\Phi_S) - \min(\Phi_S)]/\Phi_S$ sampled over 37×37 magnetic field orientations ($\theta, \phi \in [0, \pi]$) for a static $\text{FAD}^{\bullet-}/\text{W}_C^+$ pair [fixed $\mathbf{r}(t)$], including EED interactions with exchange coupling $J_0/(2\pi) = 0$ MHz, and N5 and N1 hyperfine couplings on $\text{FAD}^{\bullet-}$ and W_C^+ , respectively. (e) Harmonic driving of the interrational separation at frequency ν_d enhances the relative anisotropy, as shown for 48×100 combinations of ν_d ($1 \leq \nu_d \leq 10$ MHz) and J_0 [$-50 \leq J_0/(2\pi) \leq 50$ MHz] with EED interactions included.

where the Larmor precession frequency $\vec{\omega}_i = -\gamma_i \mathbf{B}$ is given by the product of the magnetic field $\mathbf{B} = B_0(\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$, with B_0 denoting the magnitude (approximately 50 μT for the geomagnetic field) and γ_i denoting the electron gyromagnetic ratios. The electron spin in the i th radical is denoted as $\hat{\mathbf{S}}_i$, while the nuclear spin $\hat{\mathbf{I}}_{ij}$ is coupled to the i th electron spin via hyperfine coupling tensors $\mathbf{A}_{ij}$. Additionally, we introduce time-dependent dipolar and exchange interactions between electron spins and random-field relaxation (RFR). The time-dependent part of the Hamiltonian that defines these interactions is

$$\begin{aligned}\hat{H}_1(t) &= \hat{H}_{\text{Dipolar}}(t) + \hat{H}_{\text{Exchange}}(t) + \hat{H}_{\text{RFR}}(t) \\ &= \hat{\mathbf{S}}_1 \cdot \mathbf{D}(t) \cdot \hat{\mathbf{S}}_2 - 2J(t)\hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2 \\ &\quad + \sum_{i=1,2} \sum_{j=x,y,z} \mathbf{b}_{ij}(t) \cdot \hat{\mathbf{S}}_{ij},\end{aligned}\quad (2)$$

where $\mathbf{D}(t)$ is the EED coupling tensor, $J(t)$ is the exchange coupling, and $\mathbf{b}_{ij}(t)$ denotes uncorrelated magnetic field noise of equal strength across the three Cartesian coordinates. The EED tensor and exchange coupling are proportional to $r^{-3}(t)$, and $\exp(-\beta r(t))$ with $\beta = 1.4 \text{ \AA}^{-1}$ [33], respectively. We consider harmonically driven [31] and optimally controlled [31,34] interrational modulations. The harmonic driving interrational modulation takes the form $r(t) = (\Delta_d/2)[1 - \cos(2\pi\nu_d t)] + r_0$, where ν_d is the driving frequency and the driving amplitude is set to $\Delta_d = 3 \text{ \AA}$, which was previously found to produce significant sensitivity enhancements while constituting a relatively modest motion [31]. The interrational distance of the static $\text{FAD}^{\bullet-}/\text{W}_C^+$ radical pair is specified with $r_0 = 17.2 \text{ \AA}$ informed by molecular-dynamics simulations [35]. Further details on the form of the dipolar coupling are provided in Appendix A, and how the optimally controlled modulations are obtained is provided in Appendix B, with parameters reported in Appendix C.

Following the photoinduced electron-transfer reaction that forms the candidate magnetoreceptor $\text{FAD}^{\bullet-}/\text{W}_C^+$, the initial state is an electronic singlet state, given by a spin-density operator $\hat{\rho}(0) = (\hat{P}_S/Z)$, where $\hat{P}_S$ is the singlet projection operator and $Z = Z_A Z_B$ denotes the dimension of the nuclear subspace associated with the two radicals. This initial state evolves under the total Hamiltonian $\hat{H}(t) = \hat{H}_0 + \hat{H}_1(t)$ via the master equation $\partial_t \hat{\rho}_\theta(t) = -i[\hat{H}(t), \hat{\rho}_\theta(t)] - \hat{K}(t)\hat{\rho}_\theta(t)$, where $\hat{K}(t)$ is the reaction operator comprising the reaction rate for product formation k_f and time-dependent recombination $k_b(t)$, $[\cdot, \cdot]$ indicates the commutator, and $\hat{\rho}_\theta = \hat{\rho}(t, \theta, \phi)$ signifies that the state acquires sensitivity to the magnetic field inclination through the Zeeman interaction. In this work, we restrict to $k_b(0) = k_f = 1 \mu\text{s}^{-1}$, reflecting the asymmetry

in the reaction channels from singlet and triplet states typically found in cryptochromes [10,36].

From $\hat{\rho}_\theta(t)$, the singlet population can be calculated as $p_S(t) = \text{Tr}[\hat{P}_S \hat{\rho}_\theta(t)]$, with the singlet yield defined as

$$\Phi_S = \int_0^\infty k_b(t) p_S(t) dt. \quad (3)$$

From this, the established relative anisotropy measure of sensitivity that quantifies the contrast in recombination yields over all magnetic field directions is obtained by $\Gamma = (\Phi_{S,\max} - \Phi_{S,\min})/\bar{\Phi}_S$, where $\Phi_{S,\max}$ and $\Phi_{S,\min}$ represent the maximum and minimum recombination yields and $\bar{\Phi}_S$ indicates the mean recombination yield. Figures 1(d) and 1(e) demonstrate that the modulation of interrational motion can increase yield contrast and Γ , and this has been considered in further depth in Ref. [31].

III. QUANTUM METROLOGY

Following the framework established in Ref. [26], we define the probe state as the steady state $\hat{\rho}_{ss}$ of continuous generation and recombination, evaluated from $\hat{\rho}_\theta(t)$, with specific details provided in Appendix H. As the electron spins couple to the magnetic field via the Zeeman interaction, the electronic part of $\hat{\rho}_{ss}$ encodes the intensity and directional dependence of the magnetic field. The quantum Fisher information (QFI) [37] and the associated QCRB [29] constrain the maximal precision attainable in the statistical estimation of a parameter ϑ from measurements on a quantum state $\hat{\rho}_\vartheta$. For an unbiased estimator based on N independent measurements, the bound is provided by $(1/\delta^2\vartheta) \leq NF_\vartheta \leq N\mathcal{F}_\vartheta$, where $\delta^2\vartheta$ represents the variance of the ϑ estimates, $F(\vartheta)$ is the classical Fisher information (CFI) [38,39] for any observable, and $\mathcal{F}(\vartheta)$ is the QFI. Specifically, the classical Fisher information establishes the sensitivity of the state to a particular set of measurements and takes the form

$$F_\vartheta = \sum_n p_{n,\vartheta} (\partial_\vartheta \ln p_{n,\vartheta})^2 = \sum_n \frac{(\partial_\vartheta p_{n,\vartheta})^2}{p_{n,\vartheta}}, \quad (4)$$

where $p_{n,\vartheta} = \text{Tr}[\hat{\rho}_\vartheta \hat{\Pi}_n]$ are the Born rule probabilities associated with the measurement element $\hat{\Pi}_n$, with $\sum_n \hat{\Pi}_n = \hat{I}$ and $\hat{I}$ denoting the identity operator, given that the state of the system before the measurement was $\hat{\rho}_\vartheta$. The QFI can be obtained by optimizing the CFI over all possible measurements. This framework can thus be used to analyze the quantum limit of precision in estimating a magnetic field parameter $\vartheta = \theta$ as realizable in SCRP, by choosing a set of measurements for the CFI $\Pi_n \in \{\hat{P}_S, \hat{P}_T\}$, where $\hat{P}_S$ and $\hat{P}_T = \hat{I} - \hat{P}_S$ are the singlet and triplet projection operators, respectively. This represents measurements in the magnetosensitive basis associated with chemically distinguishable outcomes, for which

spin-selective recombination reactions of the radical pair are the only anticipated mechanism in natural systems. While both the CFI and singlet yield arise from the same singlet-triplet measurement channel, the yield constitutes a time-integrated, coarse-grained observable derived from the underlying spin-state probabilities. In contrast, the CFI (which, as defined in Appendix H, is expressed in terms of $\Theta_S = \text{Tr}[\hat{P}_S \hat{\sigma}_{ss}]$, where $\hat{\sigma}_{ss}$ is the normalized steady-state spin-density operator) quantifies the information accessible through the $\{\hat{P}_S, \hat{P}_T\}$ measurement channel applied to the steady-state spin ensemble. Equality is only recovered in the static-rate limit, for which $\Phi_S \propto k_b \Theta_S$ and the yield carries the same θ dependence.

Specifically, the ratio $F_\theta/\mathcal{F}_\theta$ can be used to assess the metrological efficiency of information extraction of θ encoded in the quantum state from singlet-triplet-based measurements in an SCRP, where $F_\theta/\mathcal{F}_\theta \leq 1$ represents the degree of approach to QCRB and a value of 1 represents its saturation. Here, we have chosen to report the maximum of the ratio as a representative metric, since this point will always be present and measurable within the magnetosensitive array [Fig. 1(a)] of at least one eye, regardless of avicular orientation. Moreover, because the actual biological mechanism for magnetosensory information processing remains unknown, averaging the ratio over all orientations may obscure the intrinsic sensory precision by including magnetoreceptors not contributing to the directional estimate. In contrast, the maximum indicates the configuration in which the system most effectively exploits its available information from singlet and triplet measurements, a quantity of interest both for biological comparison and for the design of artificial radical-pair magnetometers. Nonetheless, other metrics, such as orientation-averaged ratios, are also valid and could be more appropriate for other applications. In Appendix G, we therefore report the orientation-averaged ratios; qualitatively similar trends arise under interrational motion for this alternative metric.

While the ratio $F_\theta/\mathcal{F}_\theta$ quantifies the optimality of the measurement basis relative to the quantum limit, it does not by itself ensure large absolute estimation precision. For this reason, we also report relative anisotropy values, Fisher-information magnitudes, and the corresponding angular precision $\Delta\theta = 1/\sqrt{NF_\theta}$ estimated for magnetoreceptor numbers ($N = 2 \times 10^5$ to 2×10^6) based on the energy resolution limit [40] with realistic radical-pair lifetimes in the range 1–10 μs [15,17,41,42]. Although these receptor numbers have not yet been experimentally verified, they remain biologically plausible and conservative when compared with established sensory systems such as vision, where each photoreceptor cell contains approximately 10^8 rhodopsins [43]. Our estimates are also consistent with information-theoretic analyses of magnetoreception based on photon flux and reaction yields [44]. Taken together, these comparisons demonstrate that the

observed approaches toward the QCRB can correspond to practically and biologically significant precision levels, accompanied by substantial chemical contrast.

IV. NOISE AND INTERRADICAL MODULATION

We evaluate the approach to the QCRB for a simple driven radical-pair model comprising nucleus N5, representing the dominant hyperfine coupling of $\text{FAD}^{\bullet-}/\text{W}_C^{\bullet+}$. Figure 2 shows the ratio of $F_\theta/\mathcal{F}_\theta$, which indicates how closely the system approaches the QCRB, for driving frequency ν_d against exchange coupling J_0 . Due to the approximate axial symmetry of the hyperfine interaction for this system [26], we scan over θ and set $\phi = 0$. To evaluate how noise and interrational interactions [45,46] influence metrological performance for the driven model, we consider three progressively more complex scenarios in Fig. 2: (a) exchange only, neglecting EED interactions and relaxation; (b) inclusion of EED; and (c) inclusion of both EED and RFR (with relaxation rate $\gamma = 1 \mu\text{s}^{-1}$). To maintain focus on regions of increased sensitivity, in Figs. 2(b) and 2(c), we restrict $F_\theta/\mathcal{F}_\theta$ to points where the relative anisotropy Γ is maintained or improved relative to the undriven, exchange-interaction-free reference $\Gamma(J_0 = 0, \nu_d = 0)$. Alternative noise models are compared in Appendix D, and these produce qualitatively similar results. In Fig. 2(a), where the idealized absence of dipolar and relaxation effects leads to unrealistically high baseline sensitivities, we instead restrict $F_\theta/\mathcal{F}_\theta$ to points where the

relative anisotropy does not fall below 10% of the reference. This reflects that a measurement, even if optimal, must also induce a noticeable chemical change.

Interestingly, under moderate driving ($1 \leq \nu_d \leq 100 \text{ MHz}$) within the range of biochemical plausibility as predicted from molecular-dynamics simulations [47], the system can attain 96.4% saturation of the QCRB. Moreover, with the inclusion of EED and RFR, the optimization is generally more significant, especially in the frequency range of $0 \leq \nu_d \leq 10 \text{ MHz}$, which was previously found to support relative anisotropy enhancements [31,48]. Although absolute magnitudes inevitably decrease with increasing environmental complexity, as expected for magnetoreceptive biological systems that operate at degree-level accuracy [49,50], they remain substantial. Specifically, the maximal QFI reaches $N\mathcal{F}_\theta = 32.4, 5.40$, and 0.153 ($\Delta\theta = 7.12^\circ \times 10^{-3}$ to $2.25^\circ \times 10^{-2}$, $1.74^\circ \times 10^{-2}$ to $5.51^\circ \times 10^{-2}$, and 0.104° to 0.323°), and the maximal CFI values reach $NF_\theta = 1.22, 0.708$, and 0.129 ($\Delta\theta = 3.67^\circ \times 10^{-2}$ to 0.116° , $4.81^\circ \times 10^{-2}$ to 0.152° , and 0.113° to 0.357°), for the exchange-only, with-EED, and with-RFR-and-EED scenarios, respectively. At these maximal values, the static system is vastly outperformed, with QFI smaller by 2 to 3 orders of magnitude and CFI smaller by 2 to 6 orders of magnitude. This shows that driven interrational modulations not only optimize the system in approach to the QCRB but significantly increase the absolute performance by enhancing the magnitude of precision as compared to the static scenario. Additional simulations provided in Appendix G—including full

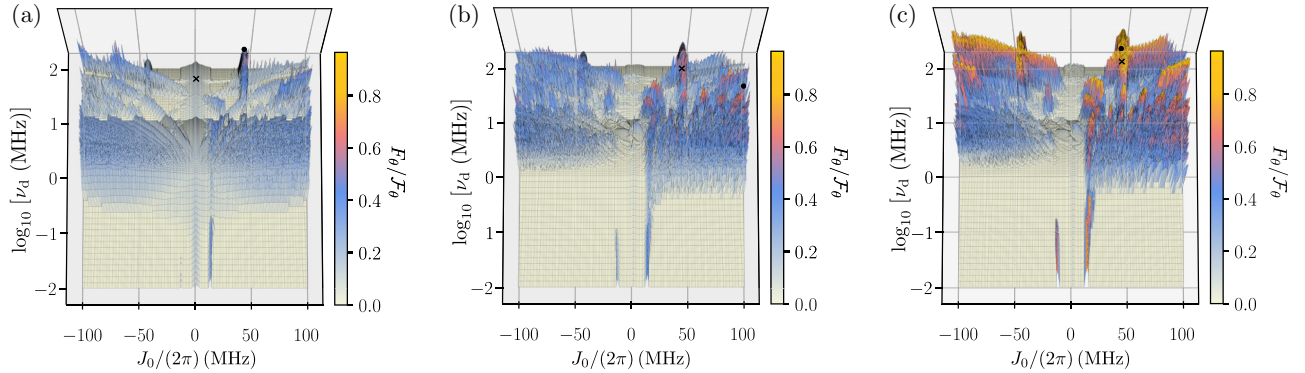


FIG. 2. Approach to the quantum Cramér-Rao bound (QCRB) under driven dynamics and increasing interaction complexity in a simple radical-pair model comprising the N5 nucleus, representing the dominant hyperfine coupling of $\text{FAD}^{\bullet-}/\text{W}_C^{\bullet+}$. The approach is assessed via the ratio of classical to quantum Fisher information $F_\theta/\mathcal{F}_\theta$, plotted as a function of 209 $\times$ 200 combinations of driving frequencies ν_d and exchange couplings J_0 spanning $0.01 \leq \nu_d \leq 100 \text{ MHz}$ and $-100 \leq J_0/(2\pi) \leq 100 \text{ MHz}$, respectively. Each point corresponds to the maximum ratio achieved over 180 magnetic field orientations ($\theta \in [0, \pi]$) at fixed $\phi = 0$. Panels show scenarios of increasing system complexity: (a) exchange only; (b) inclusion of EED interactions; (c) inclusion of both EED and random-field noise (RFR). Circles indicate closest approach to the QCRB; crosses indicate highest chemical contrast Γ . For (a), the maximum ratio 78.3% occurs at $J_0/(2\pi) = 41 \text{ MHz}$ and $\nu_d = 47 \text{ MHz}$, with $N\mathcal{F}_\theta = 0.216$ (angular precision $\Delta\theta = 2.76^\circ \times 10^{-2}$ to $8.72^\circ \times 10^{-2}$). Incorporating EED and RFR broadens the region of high ratios, reaching 96.4%, while maximum Γ values (0.456, 0.342, and 0.197) also occur at higher QCRB saturation (12%, 56.1%, and 78%). Across all these cases, maximal absolute Fisher-information magnitudes remain significant ($N\mathcal{F}_\theta \geq 0.011$), corresponding to angular precisions better than $\Delta\theta = 1.23^\circ$. Closer approaches to the QCRB with EED and RFR thus occur within practical levels of precision and reflect more efficient extraction of the available information.

plots of QFI and CFI magnitudes, relative anisotropy, and orientation-averaged values—confirm these trends and show that even field-orientation-averaged values attain ratios of up to 78.2% with mean $NF_\theta = 2.41 \times 10^{-2}$ ($\Delta\theta = 0.261^\circ\text{--}0.825^\circ$). Furthermore, in Appendix E, we demonstrate that the results persist for a wide range of modulation parameters ($0.14 \leq \beta\Delta_d \leq 14$).

V. BIOCHEMICALLY MOTIVATED SYSTEM

Moving toward a more biologically representative model of $\text{FAD}^{\bullet-}/\text{W}_C^{\bullet+}$, we include the N5 and N1 hyperfine couplings, each localized on a different radical, with EED and exchange interactions included. Figure 3(a) shows the ratio of $F_\theta/\mathcal{F}_\theta$ to indicate the approach to the QCRB, for driving frequency ν_d against exchange coupling J_0 within the range identified as beneficial in the N5 system. As axial symmetry is broken in this model system, we sampled over θ and ϕ . Here, $F_\theta/\mathcal{F}_\theta$ is displayed at all points for which there is an improvement over the static reference in the magnitudes of relative anisotropy, CFI, and QFI. We evaluated all quantities over magnetic field orientations θ and ϕ and took the maximum as representative, since a particular scanning axis through varying reaction yields could be selected or have evolved to contain the maximal point, while qualitatively similar results emerge for the average, as we show in Appendix G. In Appendix H, we provide further details on how this scheme can be viewed as a measurement of the

total electronic spin, with the CFI equivalent to the error in measuring the square of the total angular momentum $\hat{S}^2$, the relation to reaction yields, and evaluation of the QFI.

Close approaches to the QCRB are observed over a broad range of exchange couplings and driving frequencies. At the optimal point, where $F_\theta/\mathcal{F}_\theta = 89.5\%$ with $NF_\theta = 0.015$ ($\Delta\theta = 0.327^\circ\text{--}1.04^\circ$), the singlet yield Φ_S profile is plotted in Fig. 3(b) alongside its static counterpart. Driving significantly enhances both yield contrast and relative anisotropy, increasing Γ from 1.84×10^{-4} in the static system to 0.093 under driving. Unlike the static case, where yield fluctuations obscure a scanning axis through θ and ϕ , the driven yield profile exhibits a clear high-yield and low-yield point, establishing a well-defined scanning axis. Taking the points of maximum and minimum Φ_S and plotting the associated singlet probabilities p_S in Fig. 3(c) shows that the enhancement is due to the restoration of coherent singlet-triplet oscillations driven at the modulation frequency. Although driving can generally break the field-inversion symmetry, we find the effect to be minor: at the closest approach to the QCRB, the relative anisotropy differs by only 0.065% and the average yield by $1.46 \times 10^{-4}\%$ when comparing one hemisphere with the other, as shown in Appendix F. We also note that the degree of approach to the QCRB due to the inclusion of driving is broadly maintained when increasing the number of hyperfine couplings from one to two. Additional simulations provided in Appendix G—including full plots of QFI and CFI magnitudes, relative anisotropy,

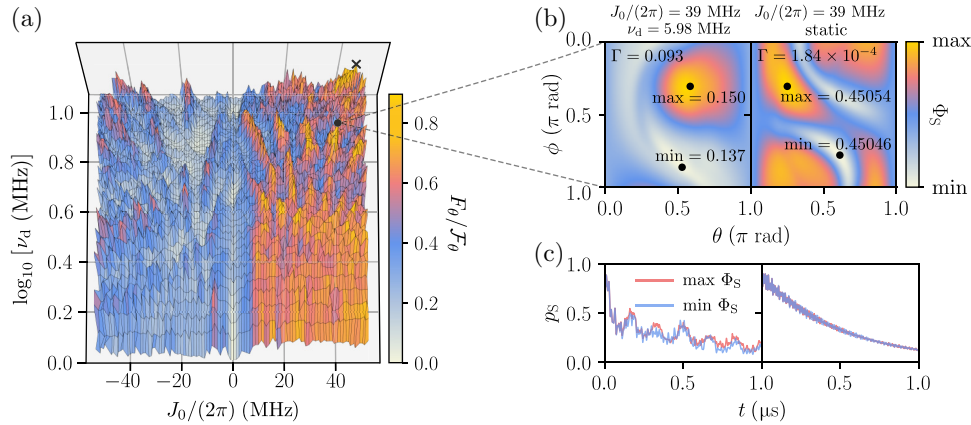


FIG. 3. Approach to the QCRB under driven dynamics in a representative radical-pair model of $\text{FAD}^{\bullet-}/\text{W}_C^{\bullet+}$ comprising N5 and N1 hyperfine couplings, electron-electron dipolar couplings, and exchange interactions. (a) The QCRB is assessed via the ratio of classical to quantum Fisher information $F_\theta/\mathcal{F}_\theta$, plotted as a function of 48×100 combinations of driving frequencies ν_d and exchange couplings J_0 spanning $1 \leq \nu_d \leq 10$ MHz and $-50 \leq J_0/(2\pi) \leq 50$ MHz, respectively. Each point corresponds to the maximum value achieved over 37×37 magnetic field orientations ($\theta, \phi \in [0, \pi]$). The closest approach to the QCRB is indicated by a circle, while the cross indicates the highest chemical contrast $\Gamma = 0.161$ with 86.8% QCRB saturation and $NF_\theta = 0.161$ (angular precision $\Delta\theta = 0.101^\circ\text{--}0.319^\circ$). (b) The singlet yield Φ_S profile for the closest approach to the QCRB of $F_\theta/\mathcal{F}_\theta = 89.5\%$, which occurs at $J_0/(2\pi) = 39$ MHz, $\nu_d = 5.98$ MHz, with $NF_\theta = 0.015$ ($\Delta\theta = 0.327^\circ\text{--}1.04^\circ$), is shown and compared to the static system, with relative anisotropy Γ indicated. (c) Singlet probability p_S over time that gives rise to the maximum and minimum singlet yields. Overall, driving provides close approaches to the QCRB, providing a larger contrast in yield and a better-defined scanning axis, originating from a restoration of coherent oscillations in the singlet probability at the driving frequency.

and orientation-averaged values—confirm these trends, showing that orientation-averaged values broadly attain ratios up to 34% with mean $N\mathcal{F}_\theta = 6.16 \times 10^{-3}$ ($\Delta\theta = 0.516^\circ$ – 1.63°).

VI. PRECISION UNDER QUANTUM CONTROL

Thus far, harmonic modulations of recombination, exchange, and electron-electron dipolar interactions have been shown to enhance magnetic field sensing through improving yield contrast, precision, and the approach to quantum limits, as assessed by the CFI and the QCRB. To assess whether more complex interrational modulation can produce further improvement, we leverage optimal quantum control [34,51–57], conditioned upon maximizing the yield contrast ($\Delta_S = \Phi_{S,\max} - \Phi_{S,\min}$) of the biologically representative $\text{FAD}^{\bullet-}/\text{W}_C^{\bullet+}$ model, to derive optimized interrational fluctuations. The latter is then used to compare to the static and harmonically driven system in Fig. 4 by generating (a) the relative anisotropy (Γ), (b) the CFI ($N\mathcal{F}_\theta$), and (c) $F_\theta/\mathcal{F}_\theta$ plotted against exchange coupling. The control results exhibit a marked enhancement across the majority of J_0 values for relative anisotropy, CFI, and QFI, reaching as high as $\Gamma = 0.863$, $N\mathcal{F}_\theta = 1.43$ ($\Delta\theta = 3.39^\circ \times 10^{-2}$ to 0.107°), and $N\mathcal{F}_\theta = 3.68$ ($\Delta\theta = 2.11^\circ \times 10^{-2}$ to $6.68^\circ \times 10^{-2}$). The harmonically driven system is generally lower, with maximum values at $\Gamma = 0.161$, $N\mathcal{F}_\theta = 0.161$ ($\Delta\theta = 0.101^\circ$ – 0.319°), and $N\mathcal{F}_\theta = 0.503$ ($\Delta\theta = 5.71^\circ \times 10^{-2}$ to 0.181°), yet it still represents a significant improvement over the maximum values of the static system at $\Gamma = 0.044$, $N\mathcal{F}_\theta = 0.036$ ($\Delta\theta = 0.213^\circ$ – 0.674°), and $N\mathcal{F}_\theta = 0.064$ ($\Delta\theta = 0.160^\circ$ – 0.507°). Furthermore, the static system for the representative scenario of $J_0/(2\pi) = 0$ MHz falls to as low as 9.11° – 28.8° , corresponding to $N\mathcal{F}_\theta = 1.98 \times 10^{-5}$. With control, approaches to the QCRB as close as

$F_\theta/\mathcal{F}_\theta = 97\%$ can be reached. In Appendix B, the absolute anisotropy (Δ_S) and magnitude of $N\mathcal{F}_\theta$ are shown.

VII. ROBUSTNESS TO BIOPHYSICAL COMPLEXITY

While relative anisotropy, and therefore chemical contrast, is known to decrease as the number of hyperfine-coupled nuclei increases in $\text{FAD}^{\bullet-}/\text{W}_C^{\bullet+}$, the factor of enhancement from interrational motion persists [31]. To test the robustness of the approach to the QCRB under increasing hyperfine complexity, we extend the model to include four hyperfine couplings (two per radical), adding the next-most-relevant hyperfine couplings N10 and H1. Figure 5 shows $F_\theta/\mathcal{F}_\theta$ over 200 driving frequencies in the range $1 \leq \nu_d \leq 100$ MHz in the presence of EED interactions with $J_0/(2\pi) = 0$ MHz, in which the insets show selected driving values of $\nu_d = 1.5, 8$, and 35 MHz where the ratio peaks, plotted over the range $-100 \leq J_0/(2\pi) \leq 100$ MHz. For computational tractability, only the orientations of θ and ϕ that maximize and minimize the static-system yield difference are scanned. Despite this minimal sampling, near saturation of the QCRB is reached (up to $F_\theta/\mathcal{F}_\theta = 93\%$), suggesting that the approach to the QCRB from interrational fluctuations is maintained as hyperfine couplings are added. To quantify the impact on absolute precision, we show in Appendix G that the four-hyperfine-coupled system exhibits a maximum $N\mathcal{F}_\theta = 0.247$ ($\Delta\theta = 8.15^\circ \times 10^{-2}$ to 0.258°), maintaining substantial precision. Extrapolating to biologically realistic regimes [58], with tens of hyperfine couplings, suggests that harmonically driven systems could yield angular precisions of the order of degrees or better. Although RFR noise would reduce the CFI by roughly an order of magnitude, structured modulations such as those identified through optimal quantum control [59] could more than compensate

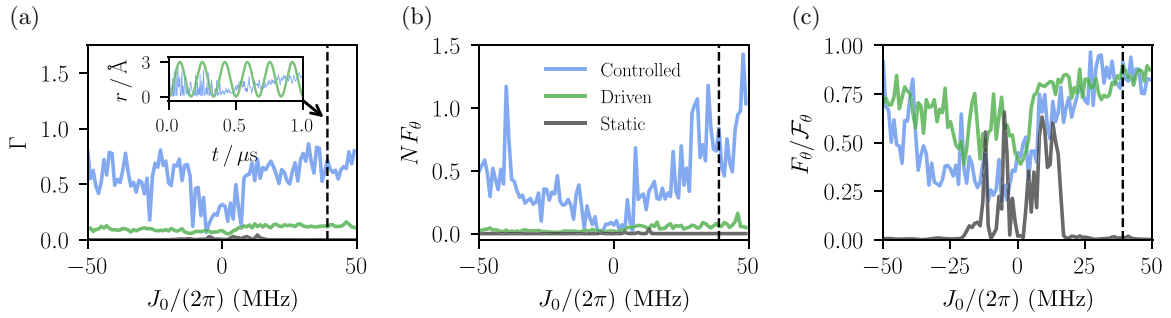


FIG. 4. Comparison of quantum-controlled interrational fluctuations that maximize yield contrast, harmonically driven fluctuations, and the static system. (a) Relative anisotropy (Γ), with the inset showing the modulated interrational distance for both the control and driven cases at the optimal exchange coupling value $J_0/(2\pi) = 39$ MHz. (b) The maximal classical Fisher information ($N\mathcal{F}_\theta$) and (c) the maximal ratios ($F_\theta/\mathcal{F}_\theta$), calculated over 37×37 combinations of $\theta, \phi \in [0, \pi]$ across the exchange coupling range $-50 \leq J_0/(2\pi) \leq 50$ MHz. For a few select points, the static system can attain comparatively high optimality due to a fortuitous exchange and dipolar cancelation (subject to strict radical placements that are in practice difficult to realize and atypical of placements in natural systems). Overall, quantum-controlled fluctuations generate the strongest enhancements, followed by the driven system, both of which significantly outperform the static system.

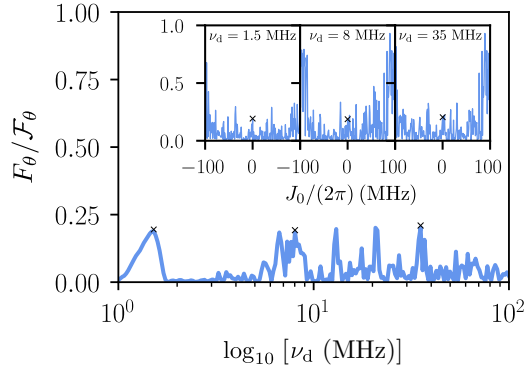


FIG. 5. Robustness of approach to the QCRB under driven dynamics of $\text{FAD}^{\bullet-}/\text{W}_C^{\bullet+}$ with four hyperfine couplings (N5 and N10 in FAD; N1 and H1 in tryptophan), electron-electron dipolar, and exchange interactions. Approach is assessed by the maximal $F_\theta/\mathcal{F}_\theta$ for the case of $J_0/(2\pi) = 0$ MHz with 200 driving frequencies in the range $1 \leq \nu_d \leq 100$ MHz, while the insets extend the exchange coupling to 200 values in the range $-100 \leq J_0/(2\pi) \leq 100$ MHz for select driving frequencies $\nu_d = 1.5, 8$, and 35 MHz. The maximal $F_\theta/\mathcal{F}_\theta$ is evaluated for orientations maximizing and minimizing the yield difference in the static case. This shows that approaches to the QCRB are maintained with increased hyperfine complexity.

for this loss. In contrast, static systems of comparable complexity perform far worse: previous analyses [26] report $NF_\theta \sim 1 \times 10^{-7}$ even without RFR, corresponding to $\Delta\theta = 128^\circ\text{--}405^\circ$. These results demonstrate that both optimality and practical precision enhancements arising from interrational motion are preserved as hyperfine interaction complexity increases.

As the model so far assumes monochromatic modulations, it is natural to ask whether more biophysically realistic dynamics can yield comparable effects. To explore this, we use estimates of dominant frequencies ω_i of motion and their relative amplitudes c_i from molecular-dynamics simulations [47], constructing a

composite modulation (up to a random phase) scaled to $\Delta_d = 3 \text{ \AA}$: $r(t) = \Delta_d[\sum_i c_i \cos(\omega_i t + \phi_i)] + r_0$, with $\phi_i \in [0, 2\pi]$. These modulations also explore shorter interrational distances and thus stronger couplings. Figure 6 shows the mean and standard error over ten phase trajectories in the model with two hyperfine couplings for (a) Γ , (b) NF_θ , and (c) $F_\theta/\mathcal{F}_\theta$, against J_0 . The results confirm that the effect broadly persists under realistic composite modulations: Γ attains approximately 0.10 ($0.05 \lesssim \Gamma \lesssim 0.15$), $F_\theta/\mathcal{F}_\theta$ reaches approximately 50%, and the maximal CFI reaches $NF_\theta = 0.0741$ ($\Delta\theta = 0.149^\circ\text{--}0.471^\circ$), while the maximal QFI is $N\mathcal{F}_\theta = 0.533$ ($\Delta\theta = 5.55^\circ \times 10^{-2}$ to 0.175°). These results show that complex, realistic fluctuations can retain much of the metrological enhancement identified under harmonic and controlled modulations, despite extending to smaller interrational distances. The path toward even greater biomimetic performance may lie in selectively inhibiting shorter-range motion and reinforcing modulation frequencies conducive to enhanced magnetosensory precision.

VIII. DISCUSSION

Experiments *in vitro* suggest a photoinduced $\text{FAD}^{\bullet-}/\text{W}_C^{\bullet+}$ radical pair [60–66], generated via electron transfer from a tryptophan residue (W_C) to a FAD cofactor, as a potential SCRP underpinning a biochemical compass sensor. An alternative candidate, $\text{FADH}^{\bullet}/\text{O}_2^{\bullet-}$, formed in the dark during flavin reoxidation with molecular oxygen, is also occasionally deliberated [67–72]. Both share the dilemma that, while simple models of their spin dynamics suggest their feasibility to underpin a compass in principle, models of realistic complexity accounting for the effects of quantum noise predict only marginal sensitivity, casting doubts on their ability to provide an effective sensory modality. To resolve this, additional enhancement mechanisms have been proposed, including exchange/dipolar cancelation [73], three-radical mechanisms [74,75], radical

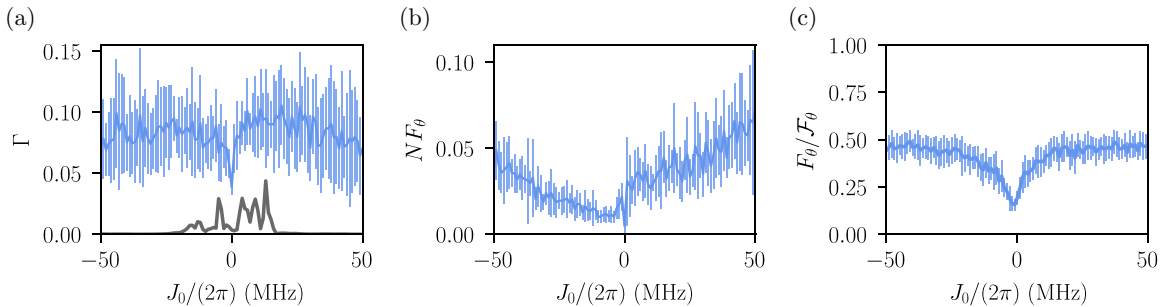


FIG. 6. Mean and standard error over ten phase trajectories of a biophysically inspired interrational modulation informed by molecular dynamics, shown for: (a) relative anisotropy (Γ), with static-system comparison; (b) classical Fisher information (NF_θ); and (c) the optimality ratio ($F_\theta/\mathcal{F}_\theta$). For each trajectory, values are calculated over 37×37 combinations of $\theta, \phi \in [0, \pi]$ across the exchange coupling range $-50 \leq J_0/(2\pi) \leq 50$ MHz, with maxima reported. The system attains up to $F_\theta/\mathcal{F}_\theta \approx 50\%$, $NF_\theta = 0.0741$ ($\Delta\theta = 0.149^\circ\text{--}0.471^\circ$), and $\Gamma \approx 0.10$, demonstrating that interrational motion continues to enhance precision, approach to the quantum Cramér-Rao bound, and chemical contrast under realistic, composite modulations.

scavenging [76], relaxation-assisted mechanisms [41,42], diffusive [77] and driven-radical motion [6,31,47], the quantum Zeno effect [78–82], and chirality-induced spin selectivity [83–89]. Various metrics, such as energy resolution limits [40], mutual information [44,90], Wigner-Yanase skew information [91], and the QFI [26,92], have been explored, relating the sensing performance to the number of magnetoreceptors, coherence, and measurement strategy.

Nevertheless, most studies of enhancement mechanisms focus on the chemical reaction yield contrast over magnetic field orientation, which does not directly establish whether the ultimate limits of quantum precision are approached, nor identify how high the ceiling of magnetometric performance lies. Our approach, by contrast, provides more direct insight into how nature may have harnessed structural adaptations from biophysics to leverage environment-assisted quantum dynamics [93–97], principles that could be exploited in tunable molecular quantum technologies [4,5] for information processing and metrology. We expect our results to generalize beyond $\text{FAD}^{\bullet-}/\text{W}_\text{C}^{\bullet+}$ to radical systems in which modulations can arise naturally or be engineered, with their underlying physical principles potentially also offering a route toward identifying biomimetic design principles for quantum sensing. Particularly promising are systems with reference-probe characteristics [98–101], such as $\text{FADH}^{\bullet-}/\text{O}_2^{\bullet-}$ or the hypothetical $\text{FAD}^{\bullet-}/\text{Z}^{\bullet}$ [98]. Future work could further investigate manipulation of structured modulations informed by molecular-dynamics simulations, where periodic motion with frequency components in the beneficial 1–10 MHz range has indeed been recently observed [47], and explore whether alternative mechanisms, such as quantum Zeno-mediated enhancements in tightly bound radicals [80], can also push performance to approach quantum limits.

IX. CONCLUSION

We have shown that interrational modulations induced by molecular motion can push the angular precision in magnetic field sensing toward the quantum limit set by the quantum Cramér-Rao bound, by increasing the metrological efficiency, quantified through the ratio $F_\theta/\mathcal{F}_\theta$, while simultaneously enhancing the absolute magnitudes of precision. This was evident both on average over the array of magnetoreceptors and, to an even greater extent, for maximal values, reflecting the best-performing subset of magnetoreceptors. Remarkably, realistic system complexities—including exchange and dipolar couplings, random-field relaxation noise, and increased hyperfine couplings—were found to augment metrological efficiency. This originates from restored singlet-triplet coherence, driven by Landau-Zener-Stückelberg-Majorana transitions [30,31,102] that

increase the chemical yield contrast and both classical and quantum Fisher information magnitudes. While even harmonic modulations are effective, modulations identified through quantum control [31,34,59,103–105], and combining harmonic components at 1–10 MHz with components resonant with hyperfine splittings, enable substantially greater precision. Such modulations, which may have emerged in cryptochromes over their 541–1000 million years of evolution from DNA photolyases [22–25], reduce the untenable angular errors predicted for the static system (exceeding 100°) to within the biological functional window of degrees [44,49,50,106], and furthermore offer scope for engineering subdegree improvements beyond what nature may have achieved.

ACKNOWLEDGMENTS

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DATA AVAILABILITY

The data that support the findings of this article are openly available [107,108].

APPENDIX A: HARMONICALLY DRIVEN INTERRADICAL MOTION

Following the approach in Ref. [31], we introduce radical motion

$$r(t) = \frac{\Delta_d}{2}[1 - \cos(2\pi\nu_d t)] + r_0, \quad (\text{A1})$$

for which the corresponding radical-motion-modulated dipolar coupling is

$$\hat{H}_{\text{Dipolar}}(r(t)) = -d(r(t))\left[3(\hat{\mathbf{S}}_1 \cdot \mathbf{v})(\hat{\mathbf{S}}_2 \cdot \mathbf{v}) - \hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2\right], \quad (\text{A2})$$

where $d(r(t)) \equiv \mu_0 g_e^2 \mu_B^2 / 4\pi r(t)^3 > 0$, with $r \equiv |\mathbf{r}|$ and constants taking their usual definitions, and $\mathbf{v} = \mathbf{r}/r$. The

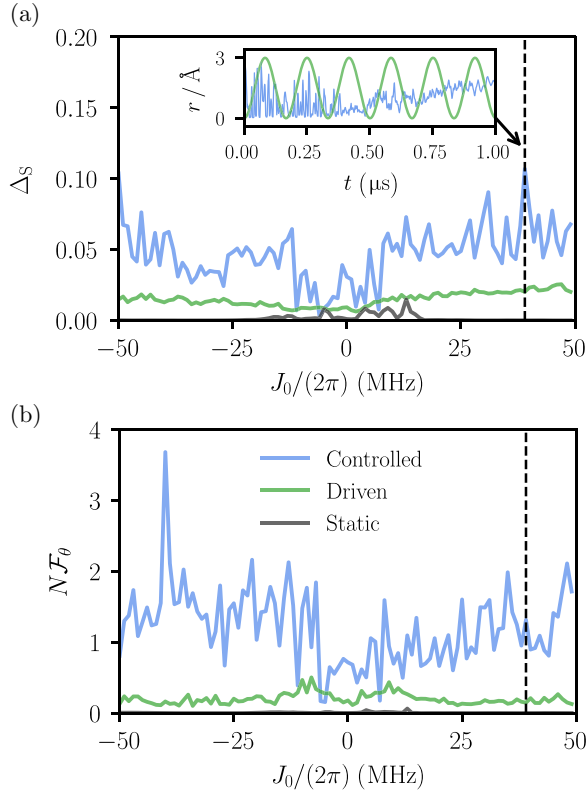


FIG. 7. Comparison of quantum-controlled interradical fluctuations that maximize yield contrast, harmonically driven fluctuations, and the static system. (a) Absolute anisotropy ($\Delta_S = \Phi_{S,\max} - \Phi_{S,\min}$), with the inset showing the modulated interradical distance for both the control and driven cases at the optimal exchange coupling value $J_0/(2\pi) = 39$ MHz, and (b) maximum quantum Fisher information $N\mathcal{F}_\theta$, calculated over 37×37 combinations of $\theta, \phi \in [0, \pi]$ across the exchange coupling range $-50 \leq J_0/(2\pi) \leq 50$ MHz, with maximum values reported. There are cases where the driven system can attain a higher approach to the QCRB than for under control, since the control target is set to maximize Δ_S (accessible to natural systems) and not the precision or optimality directly.

recombination [31] is modulated as

$$k_b(r(t)) = k_{b0} \exp[-\beta(r(t) - r_0)]. \quad (\text{A3})$$

TABLE I. Hyperfine coupling parameters in linear frequency units for nuclei in FAD of $\text{FAD}^{\bullet-}/\text{W}_C^{\bullet+}$.

Nucleus	Hyperfine interaction tensor (MHz)
N5	$\begin{bmatrix} -0.361 & -0.070 & -1.415 \\ -0.070 & -0.602 & 0.323 \\ -1.415 & 0.323 & 50.802 \end{bmatrix}$
N10	$\begin{bmatrix} 0.409 & -0.040 & -0.166 \\ -0.040 & 0.122 & -0.223 \\ -0.166 & -0.223 & 17.843 \end{bmatrix}$

TABLE II. Hyperfine coupling parameters in linear frequency units for nuclei in W_C of $\text{FAD}^{\bullet-}/\text{W}_C^{\bullet+}$.

Nucleus	Hyperfine interaction tensor (MHz)
N1	$\begin{bmatrix} 2.138 & 3.193 & -2.489 \\ 3.193 & 15.450 & -12.448 \\ -2.489 & -12.448 & 12.495 \end{bmatrix}$
H1	$\begin{bmatrix} -17.793 & 6.923 & 7.917 \\ 6.923 & -11.115 & 6.181 \\ 7.917 & 6.181 & -5.208 \end{bmatrix}$

TABLE III. Electron-electron dipolar coupling tensors in linear frequency units.

Model	EED tensor (MHz)
$\text{FAD}^{\bullet-}/\text{W}_C^{\bullet+}$	$\begin{bmatrix} 4.21 & -8.90 & 7.98 \\ -8.90 & -3.32 & 12.12 \\ 7.98 & 12.12 & -0.89 \end{bmatrix}$

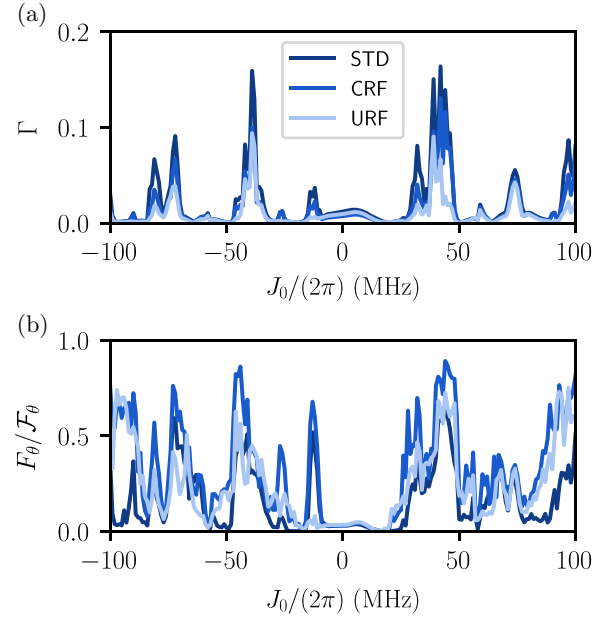


FIG. 8. Comparison of correlated random field (CRF), uncorrelated random field (URF), and singlet-triplet dephasing (STD) noise models under driven dynamics in a simple radical-pair model comprising the N5 nucleus, representing the dominant hyperfine coupling of $\text{FAD}^{\bullet-}/\text{W}_C^{\bullet+}$. (a) Relative anisotropy (Γ). (b) Approach to the quantum Cramér-Rao bound quantified via the maximal $F_\theta/\mathcal{F}_\theta$. Each data point has been calculated over 180 magnetic field orientations ($\theta \in [0, \pi]$) at fixed $\phi = 0$ for a driving frequency $\nu_d = 35$ MHz, which maximizes Γ in the reference noise model of Fig. 2.

The exchange interaction is modulated in the same functional form as

$$\hat{H}_{\text{Exchange}}(r(t)) = -2J(r(t))\hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2, \quad (\text{A4})$$

with $J(r(t)) = J_0 \exp[-\beta(r(t) - r_0)]$. Together with Eqs. (1) and (2), these define

$$\hat{H}_{\text{eff}}(r(t)) = \hat{H}(r(t)) - i \left(\frac{k_b(r(t))}{2} \hat{P}_S + \frac{k_f}{2} \hat{I} \right). \quad (\text{A5})$$

Using the above and following Ref. [26], we define the probe state as the radical pair in a steady state of continuous generation and recombination as $\hat{\rho}_{ss} = (k_0 c / Z) \tilde{G}(s=0) \hat{P}_S$, with k_0 denoting the generation rate, c the (constant) concentration of cryptochrome in the resting state, and $\tilde{G}(s)$ the Laplace transform of $\hat{G}(t)$.

APPENDIX B: OPTIMAL QUANTUM CONTROL

We discretize our control functions into piecewise-constant segments, as is standard in gradient-based approaches [109,110]. The total Hamiltonian, including the drift and control parts with interrational displacement

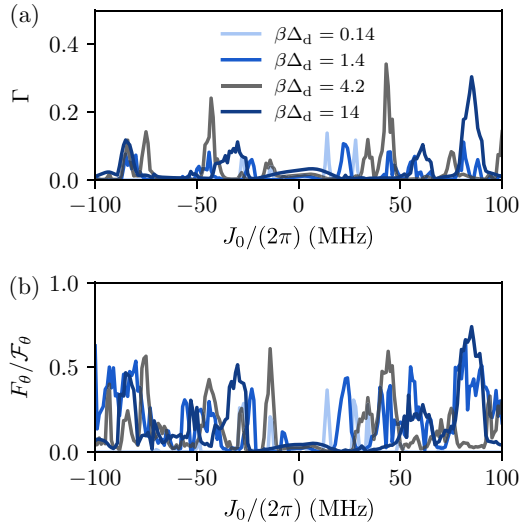


FIG. 9. Variation of the product $\beta\Delta_d$ governing the effective modulation strength of the exchange coupling against J_0 , in a simple radical-pair model comprising the N5 nucleus, representing the dominant hyperfine coupling of $\text{FAD}^{\bullet-}/\text{W}_C^{\bullet+}$, and including EED interactions. (a) Relative anisotropy (Γ). (b) Approach to the quantum Cramér-Rao bound quantified via the maximal $F_\theta/\mathcal{F}_\theta$. Each data point has been calculated over 180 magnetic field orientations ($\theta \in [0, \pi]$ at fixed $\phi = 0$) for a driving frequency $\nu_d = 36$ MHz, which maximizes Γ at the reference coupling $\beta\Delta_d = 3.4$, indicated in gray, with comparative results shown across the range $\beta\Delta_d = 0.14$ –14 in blue corresponding to $\Delta_d = 0.1$ –10 Å.

r_j constant during each interval, is

$$\hat{H}_{\text{Total}}(r_j) = \hat{H}_0 + \hat{H}_{\text{Exchange}}(r_j) + \hat{H}_{\text{Dipolar}}(r_j), \quad (\text{B1})$$

where the interrational displacement is related to the control modulation u_j as $r_j = \Delta_{\text{max}} u_j + r_0$ with $0 \leq u_j \leq 1$. Following the approach from Ref. [34], the controls are optimized to maximize the difference in yields: $\Phi_{S,\text{max}} - \Phi_{S,\text{min}}$, with min and max here referring to the magnetic field orientations of the minimal and maximal yield, respectively, in the absence of controlled distance fluctuations. To numerically solve the maximization problem, we use 1000 controls of 1 ns each, with the maximal displacement bound to $\Delta_{\text{max}} = 3$ Å, ensuring physical feasibility of the controls. With respect to the control results of Fig. 4, we consider an alternative to the relative anisotropy, the absolute anisotropy $\Delta_S = \Phi_{S,\text{max}} - \Phi_{S,\text{min}}$, and the maximum quantum Fisher information $N\mathcal{F}_\theta$ and plot these in Fig. 7.

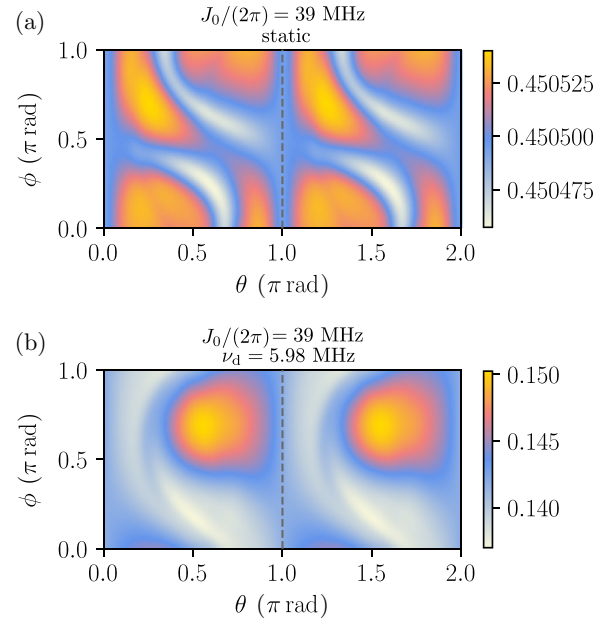


FIG. 10. The singlet yield Φ_S profile for exchange coupling $J_0/(2\pi) = 39$ MHz is shown for (a) the static system and (b) the driven system for the closest approach to the QCRB at $F_\theta/\mathcal{F}_\theta = 0.895$ in the representative radical-pair model of $\text{FAD}^{\bullet-}/\text{W}_C^{\bullet+}$ comprising the N5 and N1 hyperfine couplings and electron-electron dipolar and exchange interactions. This allows us to test the inversion asymmetry arising from the inclusion of driving when scanning separate hemispheres ($\theta = 0$ to $\theta = \pi$ vs $\theta = \pi$ to $\theta = 2\pi$). In the static case, the relative anisotropy and average yield differ by a negligible amount, reflecting the numerical accuracy of the solution approach. With the inclusion of driving, the anisotropy and average yield differ on the order of 10^{-5} and 10^{-7} , respectively. While an inversion asymmetry is thus present in the driving case, it is still several orders of magnitude smaller than relevant quantities, and the yield profiles remain similar when scanning over different hemispheres.

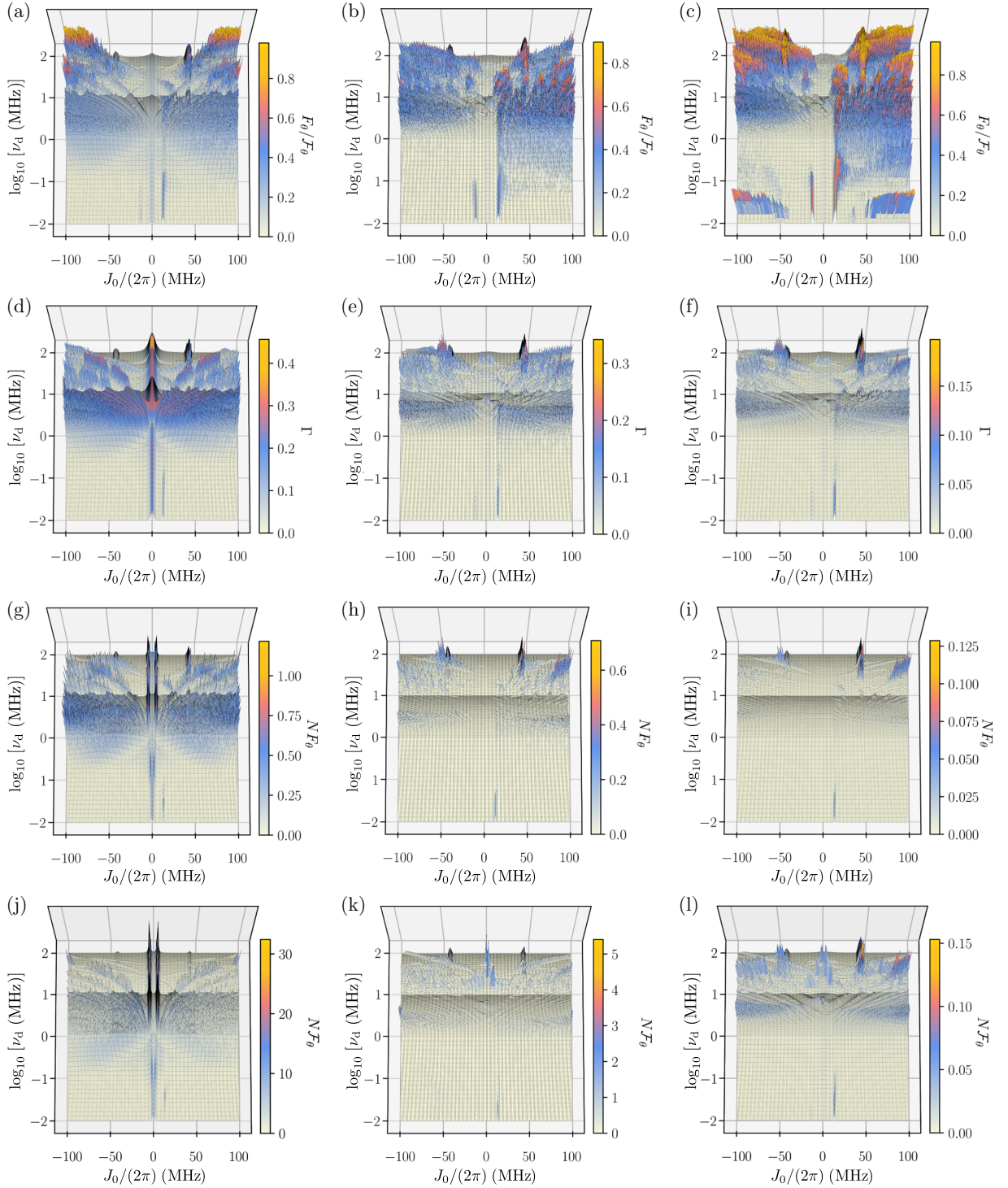


FIG. 11. Maximum values arising from driven dynamics in a simple radical-pair model comprising the N5 hyperfine interaction, representing the dominant hyperfine coupling of $\text{FAD}^{\bullet-}/\text{W}_C^{\bullet+}$, with quantities plotted as a function of 209×200 combinations of driving frequencies ν_d and exchange couplings J_0 spanning $0.01 \leq \nu_d \leq 100$ MHz and $-100 \leq J_0/(2\pi) \leq 100$ MHz, respectively. Each point (except for relative anisotropy) corresponds to the maximum achieved over 180 magnetic field orientations ($\theta \in [0, \pi]$) at fixed $\phi = 0$). Plots represent: (a)–(c) the QCRB assessed via the ratio of classical to quantum Fisher information $F_\theta/\mathcal{F}_\theta$ unfiltered against magnitude; (d)–(f) the relative anisotropy Γ ; (g)–(i) the classical Fisher information NF_θ ; and (j)–(l) the quantum Fisher information $N\mathcal{F}_\theta$. Columns show scenarios of increasing system complexity, with the first column showing exchange only, column two showing the inclusion of EED interactions, and column three showing the inclusion of both EED and RFR noise.

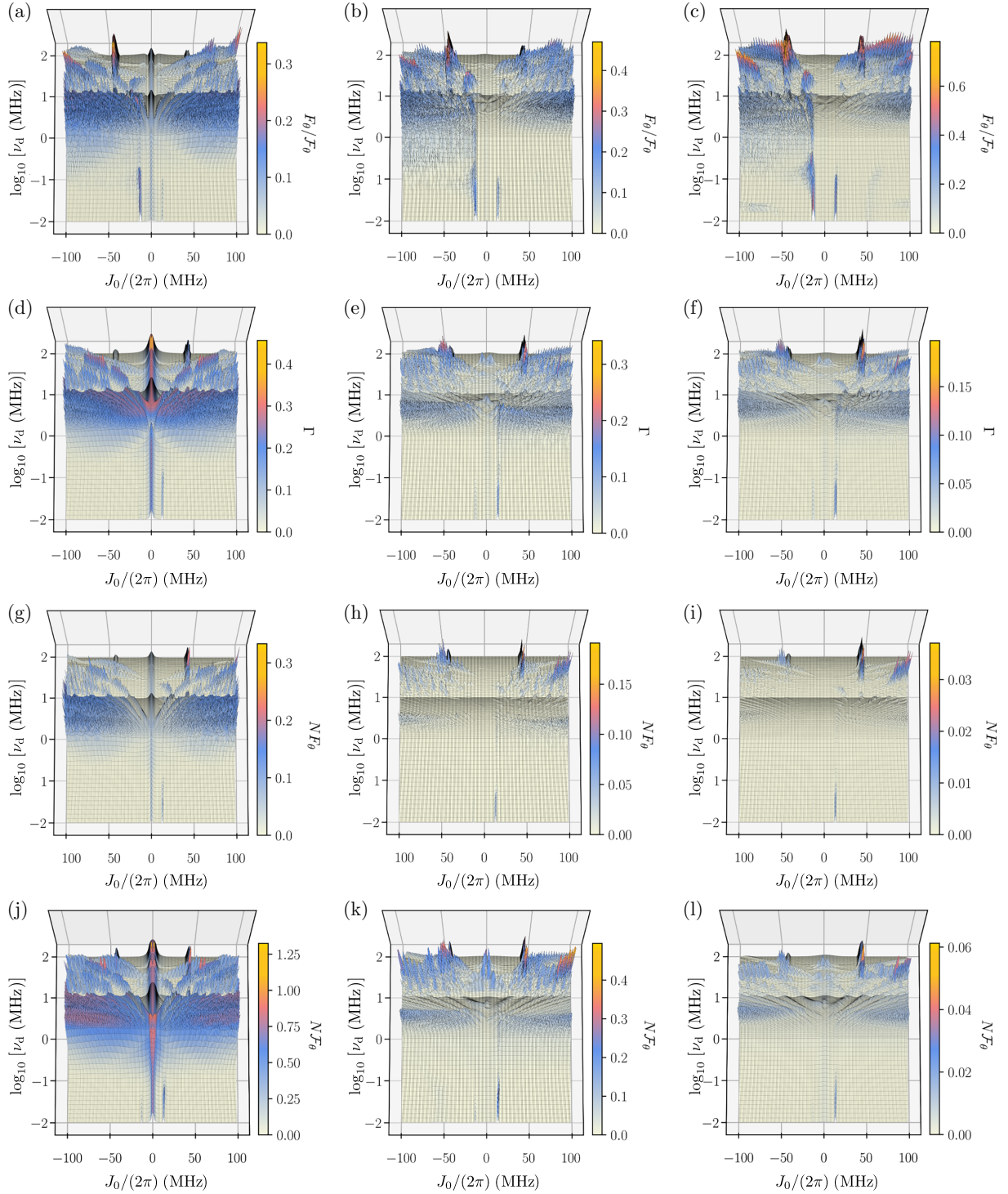


FIG. 12. Average values arising from driven dynamics in a simple radical-pair model comprising the N5 hyperfine interaction, representing the dominant hyperfine coupling of $\text{FAD}^{\bullet-}/\text{W}_C^{*+}$, with quantities plotted as a function of 209×200 combinations of driving frequencies ν_d and exchange couplings J_0 spanning $0.01 \leq \nu_d \leq 100$ MHz and $-100 \leq J_0/(2\pi) \leq 100$ MHz, respectively. Each point (except for relative anisotropy) corresponds to the average value achieved over 180 magnetic field orientations ($\theta \in [0, \pi]$) at fixed $\phi = 0$. Plots represent: (a)–(c) the QCRB assessed via the ratio of classical to quantum Fisher information $F_\theta/\mathcal{F}_\theta$ unfiltered against magnitude; (d)–(f) the relative anisotropy Γ ; (g)–(i) the classical Fisher information NF_θ ; and (j)–(l) the quantum Fisher information $N\mathcal{F}_\theta$. Columns show scenarios of increasing system complexity, with the first column showing exchange only, column two showing the inclusion of EED interactions, and column three showing the inclusion of both EED and RFR noise.

APPENDIX C: HYPERFINE AND ELECTRON-ELECTRON DIPOLAR TENSORS

Computational details for our models of $\text{FAD}^{\bullet-}/\text{W}_\text{C}^{\bullet+}$, including the hyperfine coupling tensors and the EED coupling tensor $\mathbf{D}$, are presented in Tables I–III. For $\text{FAD}^{\bullet-}/\text{W}_\text{C}^{\bullet+}$, parameters for the hyperfine coupling tensors are taken from Ref. [35] and incorporate dynamical degrees of freedom in their calculation.

APPENDIX D: ADDITIONAL NOISE MODELS

While Fig. 2 examines the influence of random-field relaxation noise on the approach to the quantum Cramér-Rao bound, and on the magnitudes of relative anisotropy and precision—chosen primarily as an indicator of the effects of noise—it is instructive to consider whether the same trends persist under alternative noise mechanisms. We thus compare three noise models: correlated random field (CRF), uncorrelated random field (URF), and singlet-triplet dephasing (STD). Their corresponding relaxation

superoperators are given by

$$\hat{R}_{\text{CRF}}[\cdot] = \gamma \sum_{\alpha=x,y,z} \mathcal{D}[\hat{S}_{1,\alpha} + \hat{S}_{2,\alpha}] \cdot, \quad (\text{D1})$$

$$\hat{R}_{\text{URF}}[\cdot] = \gamma \sum_{i=1,2} \sum_{\alpha=x,y,z} \mathcal{D}[\hat{S}_{i,\alpha}] \cdot, \quad (\text{D2})$$

$$\hat{R}_{\text{STD}}[\cdot] = \gamma \mathcal{D}[\hat{P}_\text{S}] \cdot, \quad (\text{D3})$$

where i labels the radical, x , y , and z denote Cartesian components, and $\mathcal{D}$ represents the Lindblad dissipator, $\mathcal{D}[\hat{L}]\hat{\rho} = \hat{L}\hat{\rho}\hat{L}^\dagger - \frac{1}{2}\{\hat{L}^\dagger\hat{L}, \hat{\rho}\}$, with $\{\cdot, \cdot\}$ denoting the anti-commutator. The uniform noise rate is set to $\gamma = 1 \mu\text{s}^{-1}$. Figure 8 shows Γ and $F_\theta/\mathcal{F}_\theta$ against J_0 , for $\nu_d = 35 \text{ MHz}$, the driving frequency maximizing Γ in the random-field model of Fig. 2. The results indicate that qualitatively similar behavior arises across all three noise models, confirming that the observations are not specific to the particular noise mechanism considered.

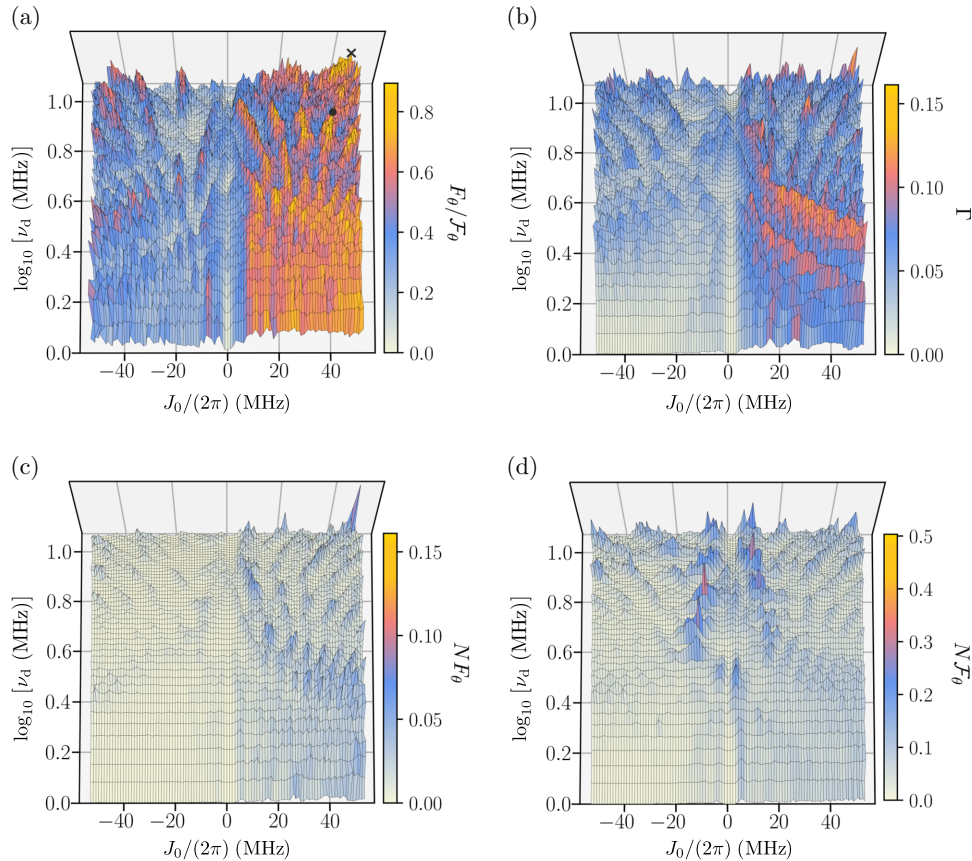


FIG. 13. Maximum values arising from driven dynamics in a representative radical-pair model of $\text{FAD}^{\bullet-}/\text{W}_\text{C}^{\bullet+}$ comprising the N5 and N1 hyperfine couplings and electron-electron dipolar and exchange interactions, with quantities plotted as a function of 48×100 combinations of driving frequencies ν_d and exchange couplings J_0 spanning $1 \leq \nu_d \leq 10 \text{ MHz}$ and $-50 \leq J_0/(2\pi) \leq 50 \text{ MHz}$, respectively. Each point (except for relative anisotropy) corresponds to the maximum value achieved over 37×37 magnetic field orientations $(\theta, \phi \in [0, \pi])$. Plots represent: (a) the QCRB assessed via the ratio of classical to quantum Fisher information $F_\theta/\mathcal{F}_\theta$; (b) the relative anisotropy Γ ; (c) the classical Fisher information NF_θ ; and (d) the quantum Fisher information $N\mathcal{F}_\theta$.

APPENDIX E: VARIATION OF MODULATION PARAMETERS

Throughout the main part of this study, we have employed a fixed set of parameters for the modulation, such as an amplitude of $\Delta_d = 3 \text{ \AA}$ and a decay constant of $\beta = 1.4 \text{ \AA}^{-1}$, as is typical for proteins [33]. These choices were motivated by biological plausibility and prior studies [31], which also indicate that the effects of driven radical motion persist across a broad range of amplitudes. Nevertheless, it is of interest to explore variations, which is realized here in terms of the product $\beta\Delta_d$, which quantifies the combined effects of modulation strength and interaction decay for the exchange interaction, while the dipolar interaction is modulated by Δ_d . Figure 9 shows Γ and $F_\theta/\mathcal{F}_\theta$ against J_0 for $\nu_d = 36 \text{ MHz}$, the driving frequency maximizing Γ for the $\text{FAD}^{\bullet-}/\text{W}_C^{\bullet+}$ pair comprising the N5 nucleus and including EED interactions. The results indicate that significant effects persist across $\beta\Delta_d = 1.4\text{--}14$, with similar peak values occurring at different J_0 , suggesting that these can be tuned alongside modulation frequencies.

APPENDIX F: INVERSION ASYMMETRY

To assess yield-inversion asymmetry that may arise due to the inclusion of interrational perturbations, we have provided full scans over the sphere in Fig. 10. This compares the static and driven models for the point of closest approach to the QCRB [$J_0/(2\pi) = 39 \text{ MHz}$, $\nu_d = 5.98 \text{ MHz}$] in the representative radical-pair model of $\text{FAD}^{\bullet-}/\text{W}_C^{\bullet+}$ comprising the N5 and N1 hyperfine couplings and electron-electron dipolar and exchange interactions (see Fig. 3). This demonstrates that similar profiles emerge when scanning over different hemispheres ($\theta = 0$ to $\theta = \pi$ vs $\theta = \pi$ to $\theta = 2\pi$), despite the presence of some small inversion asymmetry.

APPENDIX G: ANISOTROPY, AND MAXIMUM AND ORIENTATION-AVERAGED MAGNITUDES AND RATIOS

We present additional plots for the radical-pair model comprising an N5 hyperfine coupling, neglecting the EED interaction and RFR, or including them. Specifically, in

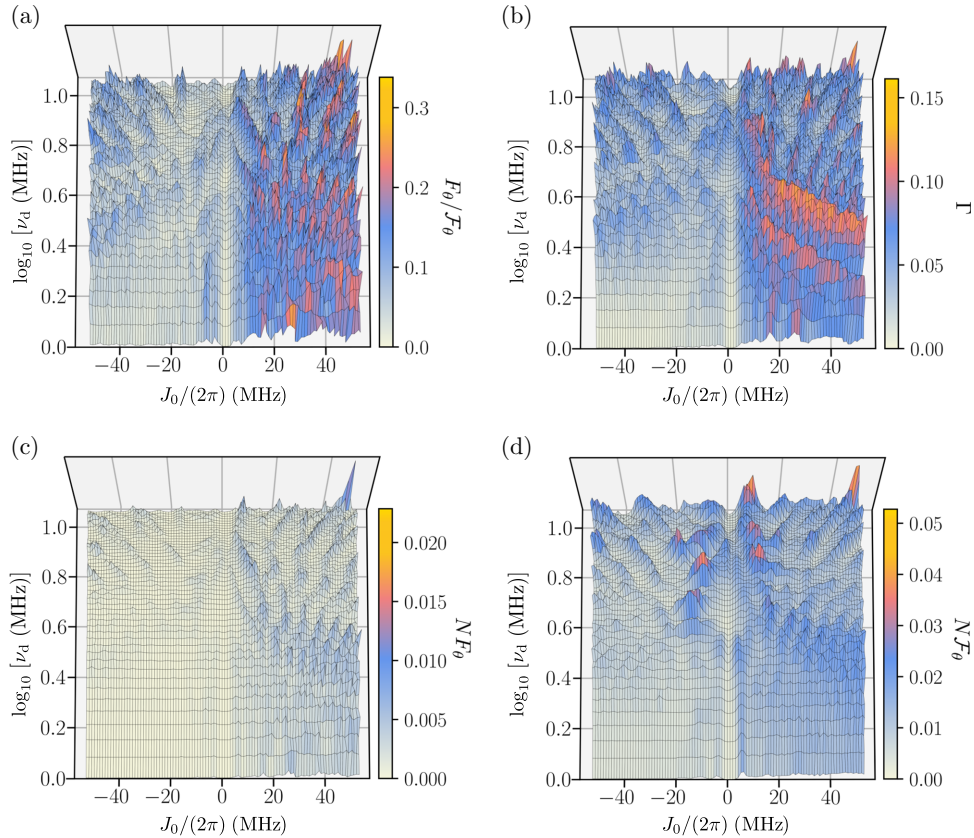


FIG. 14. Average values arising from driven dynamics in a representative radical-pair model of $\text{FAD}^{\bullet-}/\text{W}_C^{\bullet+}$ comprising the N5 and N1 hyperfine couplings and electron-electron dipolar and exchange interactions, with quantities plotted as a function of 48×100 combinations of driving frequencies ν_d and exchange couplings J_0 spanning $1 \leq \nu_d \leq 10 \text{ MHz}$ and $-50 \leq J_0/(2\pi) \leq 50 \text{ MHz}$, respectively. Each point (except for relative anisotropy) corresponds to the average value achieved over 37×37 magnetic field orientations ($\theta, \phi \in [0, \pi]$). Plots represent: (a) the QCRB assessed via the ratio of classical to quantum Fisher information $F_\theta/\mathcal{F}_\theta$; (b) the relative anisotropy Γ ; (c) the classical Fisher information $N\mathcal{F}_\theta$; and (d) the quantum Fisher information $N\mathcal{F}_\theta$.

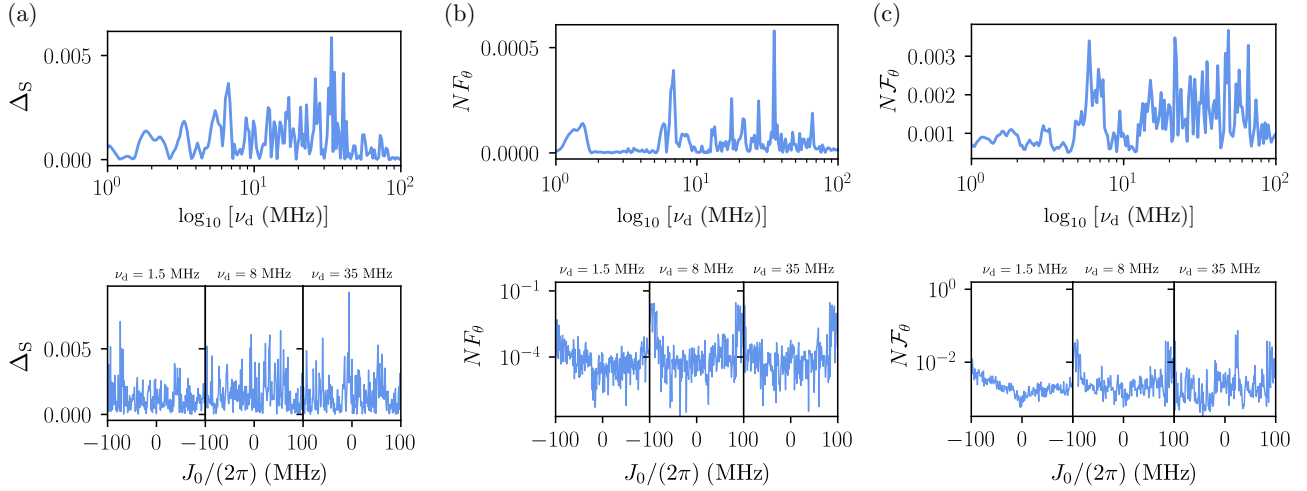


FIG. 15. Robustness of anisotropy, and the classical and quantum Fisher information under driven dynamics of $\text{FAD}^{\bullet-}/\text{W}_\text{C}^{\bullet+}$ with four hyperfine couplings (N5, N1, N10, and H1), EED, and exchange interactions. Plots represent: (a) the anisotropy; (b) the classical Fisher information NF_θ ; and (c) the quantum Fisher information $N\mathcal{F}_\theta$. The top row represents the case of EED interactions while neglecting exchange, with 200 driving frequencies in the range $1 \leq \nu_d \leq 100$ MHz, while the bottom row represents select values of driving frequencies ($\nu_d = 1.5, 8, \text{ and } 35$ MHz) that were identified as points of closest approach to the quantum Cramér-Rao bound in the main text, and the exchange coupling is extended to 200 values in the range $-100 \leq J_0/(2\pi) \leq 100$ MHz. Magnitudes have been evaluated only for orientations that maximize and minimize the yield difference of the static system, to allow for wide parameter scans of exchange coupling and driving frequency.

Figs. 11 and 12, we show the approach to the QCRB [29] assessed via $F_\theta/\mathcal{F}_\theta$, here unfiltered according to magnitude, the relative anisotropy Γ , and the magnitudes of the CFI NF_θ [38,39] and the QFI $N\mathcal{F}_\theta$ [37], calculated either for the maximum or average over orientations θ and ϕ to an external magnetic field. Similarly, Figs. 13 and 14 show these quantities for the biologically representative model of $\text{FAD}^{\bullet-}/\text{W}_\text{C}^{\bullet+}$ comprising the N5 and N1 hyperfine couplings. In Fig. 15, the magnitudes and relative anisotropy are shown under an increase of hyperfine complexity for a model comprising the N5, N1, H1, and N10 hyperfine couplings.

APPENDIX H: EXPLICIT CFI AND QFI FORMULATIONS

Extending the results of Ref. [26] to a system under time-dependent interrational motion, the CFI using Eq. (4) with measurement elements $\hat{\Pi}_n \in \{\hat{P}_\text{S}, \hat{P}_\text{T}\}$ takes the form

$$F_\theta = \frac{(\partial_\theta \text{Tr}[\hat{P}_\text{S} \hat{\sigma}_{\text{ss}}])^2}{\text{Tr}[\hat{P}_\text{S} \hat{\sigma}_{\text{ss}}]} + \frac{(\partial_\theta \text{Tr}[(\hat{I} - \hat{P}_\text{S}) \hat{\sigma}_{\text{ss}}])^2}{\text{Tr}[(\hat{I} - \hat{P}_\text{S}) \hat{\sigma}_{\text{ss}}]}$$

$$= \frac{(\partial_\theta \Theta_\text{S})^2}{\Theta_\text{S}(1 - \Theta_\text{S})}, \quad (\text{H1})$$

where $\hat{\sigma}_{\text{ss}}$ is the normalized spin-density operator and $\Theta_\text{S} = \text{Tr}[\hat{P}_\text{S} \hat{\sigma}_{\text{ss}}]$ represents the expectation value of the conditional singlet probability, which is the probability of finding the system in the singlet state, conditioned on its survival. Following the derivation in Ref. [26], it follows

that $1/\Delta^2\theta = NF_\theta$ still holds when considering the error associated with measuring the observable $\hat{O} = \hat{S}^2$ through N independent measurements; however, we note that Θ_S differs from the singlet yield Φ_S due to the time-dependent nature of the recombination rate constant, and the relation to singlet yield is only reclaimed in the limit of the static system (up to normalization).

To obtain the QFI [39], we note that $\partial_\theta p_{n,\theta} = \text{Tr}[\partial_\theta \hat{\rho}_\theta \hat{\Pi}_n] = \text{Tr}[\hat{\rho}_\theta \hat{\Pi}_n \hat{L}_\theta]$, where $\hat{L}_\theta$ is the symmetric logarithmic derivative. Maximizing the CFI over $\{\hat{\Pi}_n\}$ gives $\text{Tr}(\hat{L}_\theta^2 \hat{\rho}_\theta) = \mathcal{F}_\theta$. In the basis that diagonalizes $\hat{\rho}_\theta$, and using the spectral decomposition $\hat{\rho}_\theta = \sum_i p_i |\psi_i\rangle \langle \psi_i|$, we obtain

$$\mathcal{F}_\theta = 2 \sum_{p_i + p_j \neq 0} \frac{1}{p_i + p_j} |\langle \psi_i | \partial_\theta \hat{\rho}_\theta | \psi_j \rangle|^2. \quad (\text{H2})$$

Alternative approaches to computing the QFI are possible if $\hat{\rho}_\theta$ is invertible [111].

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