Supplementary Material: Triggering single-molecule qubit spin dynamics via non-Abelian geometric phase effects

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SUPPLEMENTARY NOTE 1: DERIVATION OF THE NON-ADIABATIC NON-ABELIAN BERRY CONNECTION

For completeness we sketch here a derivation of the non-adiabatic non-Abelian propagator for a quasi-degenerate manifold of energy eigenstates for a general quantum system. In the following, we assume the instantaneous Hilbert space of the time-dependent Hamiltonian $H(t)$ to be decomposable into a direct sum of quasi-degenerate manifolds $\mathcal{H} = \mathcal{M}_0 \oplus \cdots \oplus \mathcal{M}_P$ of instantaneous eigenstates of $H(t)$. The notion of quasi-degeneracy is here determined by the rate of change of the instantaneous energies of $H(t)$ versus their minimal spacings during the time evolution of the quantum system.

Before proceeding, we recall the conditions for adiabatic time evolution which are crucial for the observation of the Berry phase. We make the standard ansatz for the wavefunction $|\psi(t)\rangle$

$$|\psi(t)\rangle = \sum_k c_k(t) e^{-\frac{i}{\hbar}\int_0^t dt' \epsilon_k(t')} |\eta_k(t)\rangle \tag{1}$$

in terms of the time dependent coefficients $c_k(t)$, instantaneous energies $\epsilon_k(t)$ and instantaneous eigenstates $|\eta_k(t)\rangle$ (k enumerates the entire collection of instantaneous eigenstates of $H(t)$ at a given time t). The instantaneous energies and eigenstates are assumed to be known and $c_k(t)$ are to be determined. Using this ansatz in the time-dependent Schrödinger equation leads straightforwardly to the relation

$$\dot{c}_l(t) + c_l(t) \langle \eta_l(t)|\dot{\eta}_l(t)\rangle + \sum_{k\neq l} c_k(t) \langle \eta_l(t)|\dot{\eta}_k(t)\rangle = 0. \tag{2}$$

Taking a time derivative of the instantaneous "time-independent" Schrödinger equation $H(t)|\eta_k(t)\rangle = \epsilon_k(t)|\eta_k(t)\rangle$ and multiplying on the left by $\langle\eta_l(t)|$ reveals that

$$\langle \eta_l(t)|\dot{\eta}_k(t)\rangle = \frac{\langle\eta_l(t)|\dot{H}(t)|\eta_k(t)\rangle}{\epsilon_k(t) - \epsilon_l(t)} \tag{3}$$

for $\epsilon_l(t) \neq \epsilon_k(t)$ and so we have

$$\dot{c}_l(t) + c_l(t) \langle \eta_l(t)|\dot{\eta}_l(t)\rangle + \sum_{k\neq l} c_k(t) \frac{\langle\eta_l(t)|\dot{H}(t)|\eta_k(t)\rangle}{\epsilon_k(t) - \epsilon_l(t)} = 0. \tag{4}$$

If $\epsilon_l(t) \approx \epsilon_k(t)$ for some states during the time evolution of the system, the geometric terms $\langle \eta_l(t)|\dot{\eta}_k(t)\rangle$ must be considered separately. These differential equations can be decoupled insofar as the matrix elements of the variation of the Hamiltonian between different eigenstates is much smaller than their energy splitting at all times i.e. $\left|\langle\eta_l(t)|\dot{H}(t)|\eta_k(t)\rangle\right| << |\epsilon_k(t) - \epsilon_l(t)|\ \forall t$. This is the condition for adiabatic evolution within a given *quasi*-degenerate manifold. Thus for a Hamiltonian that varies linearly in time over an interval $0 \leq t \leq T$, for time-evolution restricted

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to one *quasi*-degenerate energy manifold of $H(t)$ we should have that $\frac{1}{T} << \Delta E(t)$ where $\Delta E(t)$ is the (possibly time dependent) energy spacing to the other *quasi*-degenerate manifolds.

Now we consider a given manifold $\mathcal{M}_K$ of instantaneous energy eigenstates $|\eta_k(t)\rangle$ (where k indexes the states within $\mathcal{M}_K$ only) with associated instantaneous energies $\epsilon_k(t)$. If the system is initialised as some linear combination of states from $\mathcal{M}_K$ and the time evolution is not so fast as to trigger a dynamics with states from other manifolds $\mathcal{M}_{L\neq K}$ (see above condition), then the wavefunction of the system can be expanded in eigenstates of $\mathcal{M}_K$ only, according to

$$|\psi(t)\rangle = \sum_k c_k(t)\,|\eta_k(t)\rangle\,. \tag{5}$$

Importing this expression into the time-dependent Schrödinger equation and multiplying from the left with the bra $\langle\eta_l(t)|$ gives the system of coupled differential equations

$$\dot{c}_l(t) = \frac{-i\epsilon_l(t)}{\hbar}c_l(t) - \sum_k \langle\eta_l(t)|\,\dot{\eta}_k(t)\rangle c_k(t) \tag{6}$$

involving coefficients of eigenstates from $\mathcal{M}_K$ only. Eq. (6) is solved formally by iteration to yield

$$\begin{pmatrix} c_1(t) \\ \vdots \\ c_k(t) \end{pmatrix} = \mathcal{T}e^{-\int_{t_0}^{t} dt'\left[A(t')+i\Theta(t')\right]} \begin{pmatrix} c_1(t_0) \\ \vdots \\ c_k(t_0) \end{pmatrix} \tag{7}$$

where we identify the unitary evolution operator projected onto the subspace $\mathcal{M}_K$ from time t_0 to t as

$$U(t;t_0) = \mathcal{T}e^{-\int_{t_0}^{t} dt'\left[A(t')+i\Theta(t')\right]}. \tag{8}$$

In Eq. (8), $\mathcal{T}$ is the time-ordering operator [1], $A(t)$ is the Berry connection matrix with elements $A_{ab}(t) = \langle\eta_a(t)|\,\dot{\eta}_b(t)\rangle$ and $\Theta(t)$ is the dynamical phase matrix with only non-zero diagonal elements $\Theta_{ab}(t) = \delta_{ab}\epsilon_b(t)/\hbar$.

Now, suppose that the time-dependence of $H(t)$ is resultant from the variation of d classical parameters $\boldsymbol{\lambda}(t)$ collected in the vector $\left(\lambda^1(t),\ldots,\lambda^d(t)\right)$ such that the time-dependence of the Hamiltonian, instantaneous eigenstates and energies of $\mathcal{M}_K$ can be rewritten as $H(\boldsymbol{\lambda}(t))$, $|\eta_k(\boldsymbol{\lambda}(t))\rangle$ and $\epsilon_k(\boldsymbol{\lambda}(t))$, respectively. An appropriate change of variables in the integral of Eq. (8) yields

$$U = \mathcal{P}e^{-\int_\Gamma\left[\mathcal{A}_\mu(\boldsymbol{\lambda})+i\Theta(\boldsymbol{\lambda})\frac{dt'}{d\lambda^\mu}\right]d\lambda^\mu} \tag{9}$$

where the time-ordering operator has been replaced with the path-ordering operator $\mathcal{P}$ and the line integral (with Einstein summation convention implied over the repeated index μ) is over a particular path Γ traced out in parameter space by the variation of $\boldsymbol{\lambda}$. In addition, the Berry connection matrix has been redefined with matrix elements $\mathcal{A}_{\mu,ab} = \langle\eta_a(\boldsymbol{\lambda})|\,\partial/\partial\lambda^\mu\,|\eta_b(\boldsymbol{\lambda})\rangle$ and the explicit time-dependence of the $\boldsymbol{\lambda}$'s has everywhere been suppressed. Notably, in performing this change of variables, we have assumed that the parameters $\boldsymbol{\lambda}(t')$ are invertible everywhere along the domain of integration Γ such that the derivatives $\frac{dt'}{d\lambda^\mu}$ are well-defined. For parameters which evolve linearly in time according to $\boldsymbol{\lambda}(t) = \mathbf{u} + \mathbf{v}t$ (for some time-independent d-dimensional vectors $\mathbf{u}$ and $\mathbf{v}$), it is always possible to construct such an inverse via $t = \mathbf{v}\cdot(\boldsymbol{\lambda}-\mathbf{u})/|\mathbf{v}|^2$ provided that $\mathbf{v}$ is non-singular. In this case, the derivative in Eq. 9 is simply the inverse of the linear velocity of the variation, $\left(\frac{d\lambda^\mu}{dt'}\right)^{-1}$. Note that if a particular parameter λ^ν does not vary along Γ (i.e. $\frac{d\lambda^\nu}{dt'} = 0$), it vanishes from the integral in Eq. (9) without pathology since the corresponding infinitesimal line element $d\lambda^\nu$ is also zero.

SUPPLEMENTARY NOTE 2: CALCULATION OF THE NON-ABELIAN BERRY CONNECTION MATRIX ELEMENTS FROM A MACROSCOPIC ROTATION

Here we present the explicit calculation of the non-Abelian Berry connection matrix $\mathcal{A}_\mu d\lambda^\mu$ presented in Eq. (4) of the main text. These calculations have been reported previously in the literature [2, 3] for rotations about the α and β Euler angles albeit with the third Euler angle γ set to zero.

For the ground doublet of a single-molecule magnet undergoing macroscopic rotations, the instantaneous eigenstates of the rotating crystal field Hamiltonian $H(\alpha,\beta,\gamma) = \mathcal{R}(\alpha,\beta,\gamma)H_S\mathcal{R}^\dagger(\alpha,\beta,\gamma)$ are simply $|\eta_a(\alpha,\beta,\gamma)\rangle =$

$\mathcal{R}(\alpha,\beta,\gamma)\,|\phi_a\rangle$ where $H_S\,|\phi_a\rangle = \epsilon_a\,|\phi_a\rangle$ and $\mathcal{R}(\alpha,\beta,\gamma) = \exp(-i\alpha S_z)\exp(-i\beta S_y)\exp(-i\gamma S_z)$ is the rotation operator defined in Eq. (1) of the main text. Thus, for variation of the α Euler angle, the non-Abelian Berry connection matrix element between instantaneous eigenstates $|\eta_a(\alpha,\beta,\gamma)\rangle$ and $|\eta_b(\alpha,\beta,\gamma)\rangle$ is

$$\begin{aligned}\mathcal{A}_{\alpha,ab} &= \langle\phi_a|\,\mathcal{R}^\dagger(\alpha,\beta,\gamma)\frac{\partial}{\partial\alpha}\mathcal{R}(\alpha,\beta,\gamma)\,|\phi_b\rangle \\ &= -i\,\langle\phi_a|\,e^{i\gamma S_z}e^{i\beta S_y}S_z e^{-i\gamma S_z}e^{-i\beta S_y}\,|\phi_b\rangle\,.\end{aligned} \tag{10}$$

Repeated action of the identity $e^{i\omega S_i}S_j e^{-i\omega S_i} = S_j\cos\omega - \epsilon_{ijk}S_k\sin\omega$ (where ϵ_{ijk} is the Levi-Civita tensor) leads to

$$\mathcal{A}_{\alpha,ab} = i\left(\langle\phi_a|\,S_x\,|\phi_b\rangle\sin\beta\cos\gamma - \langle\phi_a|\,S_y\,|\phi_b\rangle\sin\beta\sin\gamma - \langle\phi_a|\,S_z\,|\phi_b\rangle\cos\beta\right) \tag{11}$$

which in matrix form reads

$$\mathcal{A}_\alpha = i\left(\mathbb{S}_x\sin\beta\cos\gamma - \mathbb{S}_y\sin\beta\sin\gamma - \mathbb{S}_z\cos\beta\right). \tag{12}$$

Similarly, for variation of the β angle we have

$$\begin{aligned}\mathcal{A}_{\beta,ab} &= \langle\phi_a|\,\mathcal{R}^\dagger(\alpha,\beta,\gamma)\frac{\partial}{\partial\beta}\mathcal{R}(\alpha,\beta,\gamma)\,|\phi_b\rangle \\ &= -i\,\langle\phi_a|\,e^{i\gamma S_z}S_y e^{-i\gamma S_z}\,|\phi_b\rangle\,.\end{aligned} \tag{13}$$

where the dependence on α has vanished owing to $e^{i\alpha S_z}e^{-i\alpha S_z} = 1$. Once again employing the identity $e^{i\omega S_i}S_j e^{-i\omega S_i} = S_j\cos\omega - \epsilon_{ijk}S_k\sin\omega$ leads to

$$\mathcal{A}_{\beta,ab} = -i\left(\langle\phi_a|\,S_x\,|\phi_b\rangle\sin\gamma + \langle\phi_a|\,S_y\,|\phi_b\rangle\cos\gamma\right) \tag{14}$$

which in matrix form reads

$$\mathcal{A}_\beta = -i\left(\mathbb{S}_x\sin\gamma + \mathbb{S}_y\cos\gamma\right). \tag{15}$$

Lastly, the matrix elements for variation of the γ Euler angle are

$$\begin{aligned}\mathcal{A}_{\gamma,ab} &= \langle\phi_a|\,\mathcal{R}^\dagger(\alpha,\beta,\gamma)\frac{\partial}{\partial\gamma}\mathcal{R}(\alpha,\beta,\gamma)\,|\phi_b\rangle \\ &= -i\,\langle\phi_a|\,S_z\,|\phi_b\rangle\,.\end{aligned} \tag{16}$$

which is straightforwardly expressed in matrix form as

$$\mathcal{A}_\gamma = -i\mathbb{S}_z. \tag{17}$$

SUPPLEMENTARY NOTE 3: GEOMETRIC PROPAGATOR FOR ROTATION PATHS WITH VARYING γ ANGLE

While we have considered rotation paths in the main text with fixed γ angles, so far we have neglected contributions to the geometric propagator when γ is varied (i.e. $d\gamma \neq 0$). Owing to the simple form of the γ component of the non-Abelian gauge potential $\mathcal{A}_\gamma$, the geometric propagator about a rotation path with $d\alpha = d\beta = 0$ and $\gamma_1 \leq \gamma \leq \gamma_2$ takes the form

$$U_\gamma = e^{-\int_{\gamma_1}^{\gamma_2}\mathcal{A}_\gamma d\gamma} = e^{i(\gamma_2-\gamma_1)\mathbb{S}_z}. \tag{18}$$

For Kramers-type single-molecule magnets the $\mathbb{S}_z$ matrix has only non-zero diagonal elements and thus the propagator in Eq. (18) is Abelian. In other words, macroscopic rotation of a Kramers-type SMM about the angle γ leads, at best, to a phase acquisition of each of the ground doublet states but no population transfer. For a non-Kramers type single-molecule magnet, the $\mathbb{S}_z$ matrix can have non-zero off-diagonal matrix elements and hence a population transfer between *quasi*-degenerate ground doublet states could potentially be affected by macroscopic rotation (as demonstrated in the main text for rotations about the α angle).

SUPPLEMENTARY NOTE 4: EFFECT OF CRYSTAL FIELD SYMMETRY LOWERING AND HYPERFINE COUPLING IN $TbPc_2$

In our illustrative example of rotation-induced coherent dynamics in $TbPc_2$, we assumed that the central Tb^{3+} ion of the magnet experienced a perfect D_{4d} crystal field environment, resulting in the *quasi*-degenerate tunnel-split ground doublet $|\phi_\pm\rangle = (|m_J = -6\rangle \pm |m_J = 6\rangle)/\sqrt{2}$. To account for lowering of this symmetry, we considered a quadrupolar crystal field contribution $A_2^2\langle r^2\rangle O_2^2(\mathbf{J})$ but found little modification to the $TbPc_2$ ground doublet states unless $A_2^2\langle r^2\rangle >> A_2^0\langle r^2\rangle$. A recent multi-reference *ab initio* investigation [4] into several experimentally derived $TbPc_2$ crystal structures highlighted that symmetry lowering from ligand and/or counter ion modification, led to small asymmetries $\lesssim 7\%$ in the composition of the *quasi*-degenerate ground tunnelling states (see Tables S11-S16 of Ref. [4]).

To explore the effect of this asymmetry on the geometric phase accumulated within the ground doublet of $TbPc_2$, we consider the generalised tunnelling states $|\phi_+\rangle = \cos\frac{\kappa}{2}|m_J = -6\rangle + \sin\frac{\kappa}{2}|m_J = 6\rangle$ and $|\phi_-\rangle = -\sin\frac{\kappa}{2}|m_J = -6\rangle + \cos\frac{\kappa}{2}|m_J = 6\rangle$ as the ground doublet eigenstates of the $TbPc_2$ crystal field Hamiltonian which are separated in energy by Δ. Clearly, the $\mathbb{S}_x$ and $\mathbb{S}_y$ matrices are still zero in this basis, however the $\mathbb{S}_z$ matrix is modified from the main text according to

$$\mathbb{S}_z = J\begin{pmatrix} \cos\kappa & \sin\kappa \\ \sin\kappa & -\cos\kappa \end{pmatrix} \tag{19}$$

thus leading to the non-adiabatic propagator about a circular path on the unit sphere

$$U_{\text{circle}}^{\text{non-ad}} = e^{\frac{-iT\Delta}{2\hbar}}\begin{pmatrix} \cos\frac{\tilde{\zeta}}{2\hbar} + \frac{i(T\Delta+4\pi\hbar J\cos\beta\cos\kappa)}{\tilde{\zeta}}\sin\frac{\tilde{\zeta}}{2\hbar} & \frac{4\pi i\hbar J\sin\kappa}{\tilde{\zeta}}\cos\beta\sin\frac{\tilde{\zeta}}{2\hbar} \\ \frac{4\pi i\hbar J\sin\kappa}{\tilde{\zeta}}\cos\beta\sin\frac{\tilde{\zeta}}{2\hbar} & \cos\frac{\tilde{\zeta}}{2\hbar} - \frac{i(T\Delta+4\pi\hbar J\cos\beta\cos\kappa)}{\tilde{\zeta}}\sin\frac{\tilde{\zeta}}{2\hbar} \end{pmatrix} \tag{20}$$

where now $\tilde{\zeta} = \hbar\sqrt{8\pi^2J^2(1+\cos 2\beta) + (T\Delta/\hbar)^2 + 8\pi JT\Delta\cos\beta\cos\kappa/\hbar}$.

Following the same line of argument as in the main text, a Hadamard gate is implemented in this system if the first diagonal and off-diagonal matrix elements of Eq. (20) are equated leading to the conditions $T = (4\pi\hbar J\cos\beta/\Delta)(\sin\kappa - \cos\kappa)$ and $\tilde{\zeta}/\hbar = \sqrt{8\pi^2J^2(1+\cos 2\beta) + (T\Delta/\hbar)^2 + 8\pi JT\Delta\cos\beta\cos\kappa/\hbar} = (2n+1)\pi$. Simultaneous solution of these relations results in the polynomial

$$\cos^2\beta + \frac{\cos\kappa}{1-\sin\kappa\cos\kappa}\cos\beta - \frac{(2n+1)^2}{32J^2(1-\sin\kappa\cos\kappa)} = 0 \tag{21}$$

for $\cos\beta$. This equation has solutions

$$\cos\beta = \frac{-\cos\kappa \pm \frac{1}{2}\sqrt{\cos^2\kappa + \frac{(2n+1)^2}{8J^2}(1-\sin\kappa\cos\kappa)}}{1-\sin\kappa\cos\kappa}. \tag{22}$$

We take the positive root for $\cos\beta$ to coincide with the $\kappa = \pi/2$ result derived in the main text. Deviating from the symmetric tunnelling limit ($\kappa = \pi/2$) thus clearly requires the modulation of the canting angle and rotation time to implement a Hadamard gate via macroscopic rotation of $TbPc_2$. For the small deviations from a perfect symmetric tunnelling ground states in $TbPc_2$ that have been exposed by Pederson et al. [4], on the grounds of Eq. (22), we estimate a worst case ($\kappa = \frac{\pi}{2} \pm \frac{\pi}{25}$) correction to the canting angle of $\pm 8^\circ$ and rotation time ± 40 μs when $n = 10$.

In addition we have neglected the effect of hyperfine coupling to the $I = 3/2$ nuclear spin of ^{159}Tb as well as quadrupolar splitting of the $|m_I\rangle$ nuclear spin states due to a non-zero electric field gradient at the terbium nucleus. Both of these effects can be accounted for in our model by augmenting Eq. (16) with the terms [5]

$$H_{\text{TbPc}_2-\text{nuc}} = aJ_zI_z + P\left(I_z^2 - \frac{1}{3}I(I+1)\right) \tag{23}$$

where the hyperfine coupling parameter $a = 0.0173$ cm^{-1} and the nuclear quadrupolar splitting $P = 0.01$ cm^{-1} have been reported previously by Ishikawa et al [6]. Hyperfine coupling between the non-Kramers Tb^{3+} electronic spin and Kramers-type nuclear spin leads to a zero-field ground state doublet comprised (to a good approximation) of the $|m_J = \pm 6\rangle \otimes |m_I = \mp 3/2\rangle$ product states. The geometric propagator derived in Eq. (12) is ineffective at coupling these two states since $\mathbb{S}_x = \mathbb{S}_y = 0$ and $\mathbb{S}_z = (J-I)\sigma_z$ resulting in, at most, each state accumulating a $U(1)$ phase after rotation. The effect described in the main text can however be recovered by the application of a magnetic field $B_z = \frac{am_I}{\mu_B g_J}$ parallel to the $TbPc_2$ easy axis which also rotates simultaneously with the SMM. Such a set-up

has already been suggested for the observation of non-Abelian spin dynamics in NV centres [3] and Mn_{12} molecular nanomagnets [7]. The application of this field recovers $|\phi_{\pm}\rangle\otimes|m_I\rangle = (|m_J=-6\rangle \pm |m_J=6\rangle)/\sqrt{2}\otimes|m_I\rangle$ as an excited quasi-degenerate doublet separated from the non-degenerate ground and first few excited states by less than 0.5 K (see Figure 3a of [6]). Assuming a temperature which results in an unequal thermal population of these excited tunnelling states, the non-Abelian coherent dynamics induced by a cyclic non-adiabatic rotation is once again observed between the tunnelling ground states, as in the main text. Notably, the coherent dynamics of the non-degenerate ground and first few excited states of $TbPc_2$ in an applied field is unaffected by macroscopic rotations at speeds resonant with the tunnel splitting Δ, hence their thermal population is inconsequential to the measurement of this effect.

SUPPLEMENTARY NOTE 5: SIMULATION OF ROTATION PATH FLUCTUATIONS

To simulate stochastic fluctuations in the canting angle β originating from instrument errors (e.g. rotor jitter) during the macroscopic rotation of $TbPc_2$ studied in the main text, we pseudo-randomly sample a normal distribution with mean $\beta \approx 52^\circ$ and standard deviation σ and build up a non-constant function of β as a function of time t. In Figure 1 below we show the typical fluctuations in the β canting angle during the rotation of $TbPc_2$ about the circular path outlined in the main text. For each standard deviation, we simulate 500 rotations of the $TbPc_2$ magnet with fresh $\beta(t)$ canting angle fluctuations generated each time. We calculate the mean population and phase difference of the tunnelling states of these 500 simulations and report them in Figure 7b in the main text.

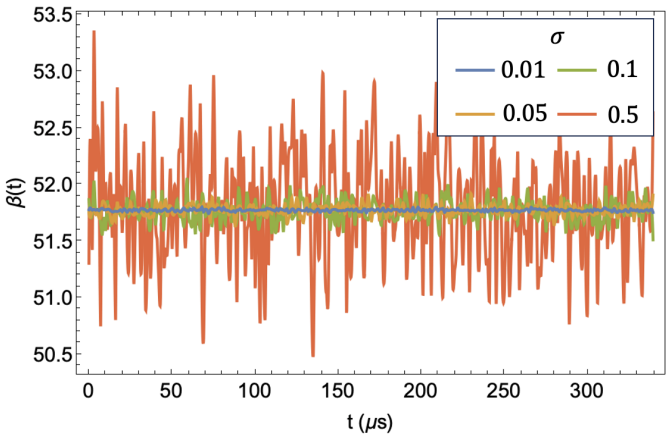

FIG. 1. Typical fluctuations of the β canting angle from sampling a normal distribution about the mean value $\beta = 51.78^\circ$ for several standard deviations σ.

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