Optimal control of NV centers

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1 Charlie - The systems model

1.1 Preliminary remarks

- In the following, we will consider the different spin-1 system (^{15}N, NV$^-$) as spin-1/2 system by considering two of the three levels as a qubit. By this reduction, one need to consider that there is a factor $\sqrt{2}$ in the control Hamiltonian. i.e. If for a spin-1 system we apply the control Hamiltonian: $u\,S_x$ then it is equivalent to applying the control Hamiltonian $\sqrt{2}\,u\,S_x$ on the spin-1/2 system.
- In the following, we will start with the energy transition written with the frequency : $\tilde{\omega}$ (in Hz). Then we define the pulsation as $\omega = 2\pi\,\tilde{\omega}$.

1.2 System 1 : NV center & nitrogen spin - Ising model

We follow here the Reference [1]. We consider the evolution of an NV center system made of the electronic spin coupled to the nitrogen nuclear spin. The dynamics of such a system is governed by the following Hamiltonian:

$$H = 2\pi\tilde{\omega}_S S_z + 2\pi\tilde{\omega}_I I_z + 2\pi\tilde{A}_\parallel S_z I_z + 2\pi\tilde{A}_\perp S_z I_x + 2\pi\tilde{u} S_x + 2\pi\tilde{v} I_x \quad (1)$$

where the S and the I operators describe respectively the electronic and nuclear spins, here considered as spin 1/2. The typical experimental values of the parameters are given in Table 1. The two control parameters are denoted $\tilde{u}(t)$ and $\tilde{v}(t)$, their respective maximum amplitudes are $\tilde{u}_{max}$ and $\tilde{v}_{max}$.

Table 1: Experimental values of the Hamiltonian parameters

Parameters	Values (MHz)
$\tilde{\omega}_S$	10^3 – 10^4
$\tilde{\omega}_I$	1
$\tilde{A}_\parallel$	S1: 1, S2: $10^{-3} - 10^{-1}$
$\tilde{A}_\perp$	S1: 1, S2: $10^{-3} - 10^{-1}$
$\tilde{u}_{max}$	10 - 100
$\tilde{v}_{max}$	1

1.3 System 2: Electron spin & weakly coupled nuclear spin

We follow here the Reference [2]. We consider the evolution of an electron spin weakly coupled to a nuclear spin. The dynamics of such a system is governed, in the secular approximation by the following Hamiltonian:

$$H = 2\pi\tilde{\omega}_S S_z + 2\pi\tilde{\omega}_I I_z + 2\pi\tilde{A}_\parallel S_z I_z + 2\pi\tilde{A}_\perp S_x I_z + 2\pi\tilde{u} S_x + 2\pi\tilde{v} I_x \tag{2}$$

where the S and the I operators describe respectively the electronic and nuclear spins, here considered as spin 1/2. The typical experimental values of the parameters are given in Table 1.

2 Charlie - Rotating frames for $A_\perp = 0$

First we consider the case $A_\perp = 0$, such the Hamiltonian of both Syst. 1 & Syst. 2 can be written the same way as, with $x = 2\pi\tilde{x}$:

$$H = \omega_S S_z + \omega_I I_z + A_\parallel S_z I_z + u S_x + v I_x. \tag{3}$$

In the following we will consider three different frames to reformulate the dynamics of the system.

2.1 Frame 1 : Double rotating frame

In this frame, we rotate the 2 spins around the z-axis at pulsation ω_S and ω_I with the Hamiltonian:

$$H_{0,dr} = \omega_S S_z + \omega_I I_z, U_{dr} = \exp(-iH_{0,dr}t) \tag{4}$$

We consider the control pulses to be of the form $u = u_0(t)\cos(\omega_S t + \varphi_S(t))$ and $v = v_0(t)\cos(\omega_I t + \varphi_I(t))$. Such that we can write, utilizing the equation (S3) the Hamiltonian in the double rotating frame as:

$$\begin{aligned} H_{dr} &= \exp(iH_{0,dr}t)\big(H - H_{0,dr}\big)\exp(-iH_{0,dr}t) && (5)\\ H_{dr} &= A_\parallel S_z I_z + u\big(S_x\cos(\omega_S t) - S_y\sin(\omega_S t)\big) + v\big(I_x\cos(\omega_I t) - I_y\sin(\omega_I t)\big). && (6)\end{aligned}$$

Such that we obtain, using the rotating wave approximation (RWA) and the trigonometric identities given in equation (S5):

$$H_{dr} = A_\parallel S_z I_z + \Omega_S\big(S_x\cos(\varphi_S) + S_y\sin(\varphi_S)\big) + \Omega_I\big(I_x\cos(\varphi_I) + I_y\sin(\varphi_I)\big) \tag{7}$$

where Ω_S and Ω_I are the Rabi frequencies of the control fields given by:

$$\begin{aligned} \Omega_S &= u_0/2, \quad \Omega_S \le u_{max}/2, && (8)\\ \Omega_I &= v_0/2, \quad \Omega_I \le v_{max}/2. && (9)\end{aligned}$$

The matrix representation of this Hamiltonian is:

$$H_{dr} = \frac{1}{2}\begin{bmatrix} A_\parallel & \Omega_I e^{-i\varphi_I} & \Omega_S e^{-i\varphi_S} & 0 \\ \Omega_I e^{i\varphi_I} & -A_\parallel & 0 & \Omega_S e^{-i\varphi_S} \\ \Omega_S e^{i\varphi_S} & 0 & -A_\parallel & \Omega_I e^{-i\varphi_I} \\ 0 & \Omega_S e^{i\varphi_S} & \Omega_I e^{i\varphi_I} & A_\parallel \end{bmatrix}$$

In the ket basis: $\{\left|\frac{1}{2}_e, \frac{1}{2}_n\right\rangle, \left|\frac{1}{2}_e, -\frac{1}{2}_n\right\rangle, \left|-\frac{1}{2}_e, \frac{1}{2}_n\right\rangle, \left|-\frac{1}{2}_e, -\frac{1}{2}_n\right\rangle\}$. For clarity, when working with $\Omega_I = 0$, we could inverse the two Hilbert spaces in the tensor product to write the matrix as :

$$H_{dr} = \frac{1}{2}\begin{bmatrix} A_\parallel & \Omega_S e^{-i\varphi_S} & 0 & 0 \\ \Omega_S e^{i\varphi_S} & -A_\parallel & 0 & 0 \\ 0 & 0 & -A_\parallel & \Omega_S e^{-i\varphi_S} \\ 0 & 0 & \Omega_S e^{i\varphi_S} & A_\parallel \end{bmatrix}$$

2.2 Frame 2 : Quadrupole rotation frame

In the previous frame each spin was rotating around an axis where the angle in the XY plan was given by φ. In this frame, the ambition is to define a new X and Y coordinate system which will follow this rotation axis. i.e $x_{new} = cos(\varphi)x + \sin(\varphi)y$. To do that we use the following transformation:

$$H_{0,qr} = \dot{\varphi}_S S_z + \dot{\varphi}_I I_z, \tag{10}$$

Such that the basis change operator is:

$$U_{qr} = \exp\big(-i\big(\varphi_S(t) - \varphi_S(0)\big)S_z - i\big(\varphi_I(t) - \varphi_I(0)\big)I_z\big) \tag{11}$$

Following the derivation given in the double rotating frame we obtain:

$$H_{qr} = A_\parallel S_z I_z - \dot{\varphi}_S S_z - \dot{\varphi}_I I_z + \Omega_S\big(S_x \cos(\varphi_S(0)) + S_y \sin(\varphi_S(0))\big) + \Omega_I\big(I_x \cos(\varphi_I(0)) + I_y \sin(\varphi_I(0))\big). \tag{12}$$

If we study a single pulse, we can actually utilize $\varphi(0) = 0$ which gives:

$$H_{qr} = A_\parallel S_z I_z - \dot{\varphi}_S S_z - \dot{\varphi}_I I_z + \Omega_S S_x + \Omega_I I_x. \tag{13}$$

To study composite pulses, or a serie of pulses then the initial value of the control pulses's phase will need to be taken into account.

The matrix representation of this Hamiltonian is:

$$H_{qr} = \frac{1}{2}\begin{bmatrix} A_\parallel - \dot{\varphi}_S - \dot{\varphi}_I & \Omega_I e^{-i\varphi_I(0)} & \Omega_S e^{-i\varphi_S(0)} & 0 \\ \Omega_I e^{i\varphi_I(0)} & -A_\parallel - \dot{\varphi}_S + \dot{\varphi}_I & 0 & \Omega_S e^{-i\varphi_S(0)} \\ \Omega_S e^{i\varphi_S(0)} & 0 & -A_\parallel + \dot{\varphi}_S - \dot{\varphi}_I & \Omega_I e^{-i\varphi_I(0)} \\ 0 & \Omega_S e^{i\varphi_S(0)} & \Omega_I e^{i\varphi_I(0)} & A_\parallel + \dot{\varphi}_S + \dot{\varphi}_I \end{bmatrix}$$

in the ket basis: $\{\left|\frac{1}{2}_e, \frac{1}{2}_n\right\rangle, \left|\frac{1}{2}_e, -\frac{1}{2}_n\right\rangle, \left|-\frac{1}{2}_e, \frac{1}{2}_n\right\rangle, \left|-\frac{1}{2}_e, -\frac{1}{2}_n\right\rangle\}$. For clarity, when working with $\Omega_I = 0$ and $\varphi(0) = 0$, we could inverse the two Hilbert spaces in the tensor product to write the matrix as :

$$H_{qr} = \frac{1}{2}\begin{bmatrix} A_\parallel - \dot{\varphi}_S & \Omega_S & 0 & 0 \\ \Omega_S & -A_\parallel + \dot{\varphi}_S & 0 & 0 \\ 0 & 0 & -A_\parallel - \dot{\varphi}_S & \Omega_S e^{-i\varphi_S} \\ 0 & 0 & \Omega_S e^{i\varphi_S} & A_\parallel + \dot{\varphi}_S \end{bmatrix}$$

2.3 Frame 3 : Interacting frame

We use here the full H_0 as the reference frame

$$H_0 = \omega_S S_z + \omega_I I_z + A_\parallel S_z I_z. \tag{14}$$

Utilizing the equation given in (S9) we have:

$$\begin{aligned} H_I =& \exp(iH_0 t)\big(H - H_0\big)\exp(-iH_0 t) && (15)\\ H_I =& u\sum_{k_I}\big(\cos\big((\omega_S + k_I A_\parallel)t\big)S_x \otimes \Pi_{k_I} - \sin\big((\omega_S + k_I A_\parallel)t\big)S_y \otimes \Pi_{k_I}\big) && (16)\\ &+ v\sum_{k_S}\big(\cos\big((\omega_I + k_S A_\parallel)t\big)\Pi_{k_S} \otimes I_x - \sin\big((\omega_I + k_S A_\parallel)t\big)\Pi_{k_S} \otimes I_y\big). && (17) \end{aligned}$$

We can then place ourself in the RWA by writing $u = u_0(t)\cos(\omega_S t + \varphi_S(t))$ and $v = v_0(t)\cos(\omega_I t + \varphi_I(t))$ we obtain:

$$\begin{aligned} H_I =& \Omega_S \sum_{k_I}\big(\cos\big(\varphi_S + k_I A_\parallel t\big)S_x \otimes \Pi_{k_I} - \sin\big(\varphi_S + k_I A_\parallel t\big)S_y \otimes \Pi_{k_I}\big) && (18)\\ &+ \Omega_I \sum_{k_S}\big(\cos\big(\varphi_I + k_S A_\parallel t\big)\Pi_{k_S} \otimes I_x - \sin\big(\varphi_I + k_S A_\parallel t\big)\Pi_{k_S} \otimes I_y\big). && (19) \end{aligned}$$

The matrix representation of this Hamiltonian is:

$$H_I = \frac{1}{2}\begin{bmatrix} 0 & \Omega_I e^{-i(\varphi_I + A_\parallel t)} & \Omega_S e^{-i(\varphi_S + A_\parallel t)} & 0 \\ \Omega_I e^{i(\varphi_I + A_\parallel t)} & 0 & 0 & \Omega_S e^{-i(\varphi_S - A_\parallel t)} \\ \Omega_S e^{i(\varphi_S + A_\parallel t)} & 0 & 0 & \Omega_I e^{-i(\varphi_I - A_\parallel t)} \\ 0 & \Omega_S e^{i(\varphi_S - A_\parallel t)} & \Omega_I e^{i(\varphi_I - A_\parallel t)} & 0 \end{bmatrix}$$

in the ket basis: $\{\left|\frac{1}{2}_e, \frac{1}{2}_n\right\rangle, \left|\frac{1}{2}_e, -\frac{1}{2}_n\right\rangle, \left|-\frac{1}{2}_e, \frac{1}{2}_n\right\rangle, \left|-\frac{1}{2}_e, -\frac{1}{2}_n\right\rangle\}$. For clarity, when working with $\Omega_I = 0$, we could inverse the two Hilbert spaces in the tensor product to write the matrix as :

$$H_I = \frac{1}{2}\begin{bmatrix} 0 & \Omega_S e^{-i(\varphi_S + A_\parallel t)} & 0 & 0 \\ \Omega_S e^{i(\varphi_S + A_\parallel t)} & 0 & 0 & 0 \\ 0 & 0 & 0 & \Omega_S e^{i(\varphi_S - A_\parallel t)} \\ 0 & 0 & \Omega_S e^{i(\varphi_S - A_\parallel t)} & 0 \end{bmatrix}$$

3 Charlie - Grape

For this section we discretize, we want to fine a control that minimize a given cost function (C). Following the calculation of the gradient of the cost, for both *state to state* or *target operator*, we need to compute the partial derivate: $\partial_{u_n} U_n$. In the following we derivate this term for both Frame 1 and Frame 2.

3.1 Derivation of evolution operator

3.1.1 Frame 1: Double rotating frame

in the double rotating frame the Hamiltonian is given by:

$$H_{dr} = A_\parallel S_z I_z + \Omega_S\big(S_x \cos(\varphi_S) + S_y \sin(\varphi_S)\big) + \Omega_I\big(I_x \cos(\varphi_I) + I_y \sin(\varphi_I)\big) \tag{20}$$

such that for the time step $n \in [0, N-1]$, we can write the evolution operator as:

$$U_n = \exp(-iH_{dr,n}\Delta t). \tag{21}$$

Using the split operator method, which states that if Δt is small enough all the operators commute with one another, we can easily compute the derivate of U_n:

$$\begin{aligned} \partial_{\Omega_{S,n}} U_n &= -i\Delta t(\cos(\varphi_{S,n})S_x + \sin(\varphi_{S,n})S_y)U_n. &(22)\\ \partial_{\varphi_{S,n}} U_n &= -i\Delta t\Omega_{S,n}(\cos(\varphi_{S,n})S_y - \sin(\varphi_{S,n})S_x)U_n, &(23)\\ \partial_{\Omega_{I,n}} U_n &= -i\Delta t(\cos(\varphi_{I,n})I_x + \sin(\varphi_{I,n})I_y)U_n, &(24)\\ \partial_{\varphi_{I,n}} U_n &= -i\Delta t\Omega_{I,n}(\cos(\varphi_{I,n})I_y - \sin(\varphi_{I,n})S_x)U_n. &(25)\\ & &(26) \end{aligned}$$

These formula can then be simply implemented in the calculation of the gradient according to your cost function.

3.1.2 Frame 2: Quadrupole rotating frame

in the quadrupole rotating frame the Hamiltonian is given by:

$$H_{qr} = A_\parallel S_z I_z - \dot{\varphi}_S S_z - \dot{\varphi}_I I_z + \Omega_S\big(\cos(\varphi_S(0))S_x + \sin(\varphi_S(0))S_y\big) + \Omega_I\big(\cos(\varphi_I(0))I_x + \sin(\varphi_I(0))I_y\big). \tag{27}$$

such that for the time step $n \in [0, N-1]$, we can write the evolution operator as:

$$U_n = \exp(-iH_{qr,n}\Delta t). \tag{28}$$

Using the split operator method, which states that if Δt is small enough all the operators commute with one another, we can easily compute the derivate of U_n:

$$\begin{aligned} \partial_{\Omega_{S,n}} U_n &= -i\Delta t(\cos(\varphi_S(0))S_x + \sin(\varphi_S(0))S_y)U_n. &(29)\\ \partial_{\dot{\varphi}_{S,n}} U_n &= -i\Delta t(-S_z)U_n &(30)\\ \partial_{\Omega_{I,n}} U_n &= -i\Delta t(\cos(\varphi_I(0))I_x + \sin(\varphi_I(0))I_y)U_n, &(31)\\ \partial_{\dot{\varphi}_{I,n}} U_n &= -i\Delta t(-I_z)U_n. &(32) \end{aligned}$$

We keep the $-i\Delta t$ form, such that we keep the same sign at the end of the gradient derivation. These formula can then be simply implemented in the calculation of the gradient according to your cost function.

3.2 $\Omega_I = 0, \varphi_S(0) = 0$

With only the use of the mw pulse (acting on the electron spin, Ω_S) and studying a single pulse ($\varphi(0) = 0$), we can work in two (2,2) matrices instead of a (4,4) matrices. To do so we define a target evolution operator $U_{t,\pm}$ for each subspace of the nuclear spin state $m_I = \pm\frac{1}{2}$.

3.2.1 Frame 1 (dr)

In the case of the double rotating frame, for each subspace we define the Hamiltonian:

$$H_{dr,\pm} = \pm\frac{A_\parallel}{2} S_z + \Omega_S\big(S_x \cos(\varphi_S) + S_y \sin(\varphi_S)\big). \tag{33}$$

Which leads to :

$$\begin{aligned}
\partial_{\Omega_{S,n}} U_{n,\pm} &= -i\Delta t(\cos(\varphi_{S,n})S_x + \sin(\varphi_{S,n})S_y)U_{n,\pm} &&= -i\Delta t H_n^\Omega U_{n,\pm}. && (34)\\
\partial_{\varphi_{S,n}} U_{n,\pm} &= -i\Delta t \Omega_{S,n}(\cos(\varphi_{S,n})S_y - \sin(\varphi_{S,n})S_x)U_{n,\pm} &&= -i\Delta t H_n^\varphi U_{n,\pm}, && (35)\\
& && && (36)
\end{aligned}$$

with $U_{n,\pm} = \exp(-i\Delta t H_{dr,\pm})$. If we define the cost function as:

$$C = 1 - \frac{1}{4}(|\mathrm{tr}(\mathrm{U}_{t,+}^\dagger \mathrm{U}_+(t_f))|^2 + |\mathrm{tr}(\mathrm{U}_{t,-}^\dagger \mathrm{U}_-(t_f))|^2). \tag{37}$$

Then we can write:

$$\begin{aligned}
\delta\Omega_{S,n} &= \epsilon\Im\big(\mathrm{tr}(\mathrm{U}_+^\dagger(t_f)\mathrm{U}_{t,+})\mathrm{tr}(\mathrm{V}_+^\dagger(t_n)\mathrm{H}_n^\Omega \mathrm{U}_+(t_n)) + \mathrm{tr}(\mathrm{U}_-^\dagger(t_f)\mathrm{U}_{t,-})\mathrm{tr}(\mathrm{V}_-^\dagger(t_n)\mathrm{H}_n^\Omega \mathrm{U}_-(t_n))\big), && (38)\\
\delta\varphi_{S,n} &= \epsilon\Im\big(\mathrm{tr}(\mathrm{U}_+^\dagger(t_f)\mathrm{U}_{t,+})\mathrm{tr}(\mathrm{V}_+^\dagger(t_n)\mathrm{H}_n^\varphi \mathrm{U}_+(t_n)) + \mathrm{tr}(\mathrm{U}_-^\dagger(t_f)\mathrm{U}_{t,-})\mathrm{tr}(\mathrm{V}_-^\dagger(t_n)\mathrm{H}_n^\varphi \mathrm{U}_-(t_n))\big). && (39)
\end{aligned}$$

with $u_n^{(k+1)} = u_n^{(k)} + \delta u_n^{(k)}$, $V_\pm(t_n) = U_n^\dagger ... U_{N-1}^\dagger U_{t,\pm}$ and $U_\pm(t_n) = U_{n-1}...U_0$.

3.2.2 Frame 2 (qr)

In the case of the quadrupole rotating frame, for each subspace we define the Hamiltonian:

$$H_{qr,\pm} = \pm\frac{A_\parallel}{2} S_z - \dot\varphi_S S_z + \Omega_S S_x \tag{40}$$

Which leads to :

$$\begin{aligned}
\partial_{\Omega_{S,n}} U_{n,\pm} &= -i\Delta t S_x U_{n,\pm}. && (41)\\
\partial_{\dot\varphi_{S,n}} U_{n,\pm} &= -i\Delta t(-S_z)U_{n,\pm}. && (42)
\end{aligned}$$

with $U_{n,\pm} = \exp(-i\Delta t H_{qr,\pm})$. If we define the cost function as:

$$C = 1 - \frac{1}{4}(|\mathrm{tr}(\mathrm{U}_{t,+}^\dagger \mathrm{U}_+(t_f))|^2 + |\mathrm{tr}(\mathrm{U}_{t,-}^\dagger \mathrm{U}_-(t_f))|^2). \tag{43}$$

Then we can write:

$$\begin{aligned}
\delta\Omega_{S,n} &= \epsilon\Im\big(\mathrm{tr}(\mathrm{U}_+^\dagger(t_f)\mathrm{U}_{t,+})\mathrm{tr}(\mathrm{V}_{n,+}^\dagger \mathrm{S_x} \mathrm{U}_{n,+}) + \mathrm{tr}(\mathrm{U}_-^\dagger(t_f)\mathrm{U}_{t,-})\mathrm{tr}(\mathrm{V}_{n,-}^\dagger \mathrm{S_x}\mathrm{U}_{n,-})\big), && (44)\\
\delta\dot\varphi_{S,n} &= -\epsilon\Im\big(\mathrm{tr}(\mathrm{U}_+^\dagger(t_f)\mathrm{U}_{t,+})\mathrm{tr}(\mathrm{V}_{n,+}^\dagger \mathrm{S_z} \mathrm{U}_{n,+}) + \mathrm{tr}(\mathrm{U}_-^\dagger(t_f)\mathrm{U}_{t,-})\mathrm{tr}(\mathrm{V}_{n,-}^\dagger \mathrm{S_z}\mathrm{U}_{n,-})\big). && (45)
\end{aligned}$$

with $u_n^{(k+1)} = u_n^{(k)} + \delta u_n^{(k)}$, $V_\pm(t_n) = U_n^\dagger ... U_{N-1}^\dagger U_{t,\pm}$ and $U_\pm(t_n) = U_{n-1}...U_0$.

4 Old notes from Dominique

We consider control pulses of the form $u_x = u_0(t)\cos(\omega_S t + \varphi(t))$ and $v = v_0(t)\cos(\omega_I t + \psi(t))$, and we

In the doubly rotating frame at frequencies ω_S and ω_I defined by the unitary transformation $U_{rot} = \exp[-i(2\pi\omega_S S_z + 2\pi\omega_I I_z)]$, we obtain within the rotating wave approximation that the Hamiltonian can be expressed as

$$H_{rot} = 2\pi A_\parallel S_z I_z + 2\pi A_\perp S_x I_x + 2\pi A_\perp S_y I_y + 2\pi u_x S_x + 2\pi u_y S_y + 2\pi v_x I_x + 2\pi v_y I_y$$

with

$$u_x^2 + u_y^2 \leq u_{max}^2;\ v_x^2 + v_y^2 \leq v_{max}^2$$

We also recall some notations in the control of a spin 1/2 particle of the form

$$S_z \rightarrow_{\frac{\pi}{2} S_y} S_x$$

This transformation of a spin 1/2 particle corresponds to a rotation of angle $\frac{\pi}{2}$ along the y- direction. Explicitly, it means that we start for the density matrix $\rho_i = \frac{1}{2} + S_z$ and that the corresponding evolution operator is $U = \exp\left[-i\frac{\pi}{2}S_y\right]$. Units are such that $\hbar = 1$. We have

$$U = \frac{1}{\sqrt{2}}(1 - i\sigma_y)$$

and we deduce that

$$S_x = U S_z U^\dagger$$

that is $\rho_f = \frac{1}{2} + S_x$.

5 Owen – Double-Rotating frame, seen as two uncoupled spins

We consider in this part the double-rotating frame Hamiltonian H_{dr} with $\Omega_I = 0$ presented in Subsec. 2.1

$$H_{dr} = \frac{1}{2}\begin{bmatrix} A_\parallel & \Omega_S e^{-i\varphi_S} & & \\ \Omega_S e^{i\varphi_S} & -A_\parallel & & \\ & & -A_\parallel & \Omega_S e^{-i\varphi_S} \\ & & \Omega_S e^{i\varphi_S} & A_\parallel \end{bmatrix} \tag{46}$$

where the zeros of the matrix have been left blank for the sake of clarity. We use for control the off-diagonal terms of the matrix

$$\Omega(t) = \Omega_S(t) e^{i\varphi_S(t)} = \Omega_x(t) + i\Omega_y(t)$$

with the constraint that Ω_S is limited in amplitude: $\Omega_S \leq \Omega_0$.

We can modelize this Hamiltonian as two independant pseudo-spin Hamiltonian (labeled 1 and 2), such that H_{dr} can be seen as their tensor product:

$$H_k = \frac{1}{2}\begin{pmatrix} A_k & \Omega^* \\ \Omega & -A_k \end{pmatrix} = \frac{1}{2}\left(A_k\sigma_z + \Omega_x\sigma_x + \Omega_y\sigma_y\right) \quad ; \quad A_{1,2} = \pm A_\parallel \tag{47}$$

with $\vec{\sigma}$ the Pauli vector. One may note that the spin 1 correspond to the nuclear spin, when the electronic spin is $\frac{1}{2}$ (basis $\left\{|0\rangle = \left|\frac{1}{2}_e, \frac{1}{2}_n\right\rangle, |1\rangle = \left|\frac{1}{2}_e, -\frac{1}{2}_n\right\rangle\right\}$), while spin 2 is the nuclear spin with the electronic spin $-\frac{1}{2}$ (basis $\left\{|0\rangle = \left|-\frac{1}{2}_e, \frac{1}{2}_n\right\rangle, |1\rangle = \left|-\frac{1}{2}_e, -\frac{1}{2}_n\right\rangle\right\}$).

Our goal is to create a 2-qubits quantum gate in the $SU(4)$ space created by the $\mathfrak{su}(4)$ Hamiltonian H_{dr}. With this modelization as two independant $\mathfrak{su}(2)$ Hamiltonians $H_{1,2}$ and using the Cayley-Klein parameters, this can be seen as a population transfer for each spin from an initial state $\left|\psi_1^0\right\rangle \otimes \left|\psi_2^0\right\rangle = |0\rangle \otimes |0\rangle$ to a final state $\left|\psi_1^f\right\rangle \otimes \left|\psi_2^f\right\rangle$, in minimum time. The Pontryagin Hamiltonian H_P of the system can be written as

$$H_P = \sum_k \mathrm{Im}\left\langle\chi_k\right| H_k \left|\psi_k\right\rangle \tag{48}$$

where $|\chi_k\rangle$ is the adjoint state of ψ_k, which follows the dynamics $\dot{\langle\chi_k|} = -2\partial_{|\psi_k\rangle} H_P = i\left\langle\chi_k\right| H_k$. Its initial state is noted $\left\langle\chi_k^0\right| = (a_k, b_k)$. In the case where $|\psi(t)\rangle$ is optimal, since the final time is not fixed, we have $H_P = 1$.

Let's set the vectors $\vec{R_k} = \frac{1}{2}\mathrm{Im}\left\langle\chi_k\right|\vec{\sigma}\left|\psi_k\right\rangle$ of $\mathbb{R}^3$, which follow the dynamics

$$\dot{\vec{R}}_k = \frac{1}{2}\mathrm{Im}\left(i\left\langle\chi_k\right|\left[H_k, \vec{\sigma}\right]\left|\psi_k\right\rangle\right) \tag{49a}$$

$$= \begin{pmatrix} & -A_k & \Omega_y \\ A_k & & -\Omega_x \\ -\Omega_y & \Omega_x & \end{pmatrix}\vec{R}_k \tag{49b}$$

We also define their sum $\vec{s} = \vec{R}_1 + \vec{R}_2$ and difference $\vec{d} = \vec{R}_1 - \vec{R}_2$, with the associated dynamics

$$\dot{\vec{s}} = \begin{pmatrix} & -A_\parallel & \\ A_\parallel & & \\ & & \end{pmatrix} \vec{d} + \begin{pmatrix} & & \Omega_y \\ & & -\Omega_x \\ -\Omega_y & \Omega_x & \end{pmatrix} \vec{s} \tag{50a}$$

$$\dot{\vec{d}} = \begin{pmatrix} & -A_\parallel & \\ A_\parallel & & \\ & & \end{pmatrix} \vec{s} + \begin{pmatrix} & & \Omega_y \\ & & -\Omega_x \\ -\Omega_y & \Omega_x & \end{pmatrix} \vec{d} \tag{50b}$$

One may note that $s^2 + d^2 = \vec{s}^2 + \vec{d}^2$ is a constant of motion, since the ${R_k}^2 = \vec{R}_k^{\,2}$ are constants of motion.

The initial states $\vec{R}_k^0$, $\vec{s}^0$ and $\vec{d}^0$ are given by

$$\vec{R}_k^0 = \frac{1}{2}\,\mathrm{Im}\begin{pmatrix} b_k \\ ib_k \\ a_k \end{pmatrix} \quad ; \quad \vec{s}^0 = \frac{1}{2}\,\mathrm{Im}\begin{pmatrix} b_1 + b_2 \\ i(b_1 + b_2) \\ a_1 + a_2 \end{pmatrix} \quad ; \quad \vec{d}^0 = \frac{1}{2}\,\mathrm{Im}\begin{pmatrix} b_1 - b_2 \\ i(b_1 - b_2) \\ a_1 - a_2 \end{pmatrix} \tag{51}$$

Using those new notations, the Pontryagin Hamiltonian writes

$$H_P = A_\parallel d_z + \Omega_x s_x + \Omega_y s_y \tag{52}$$

If we consider a regular solution (i.e. $\Omega_s = \Omega_0 \equiv 1$ without loss of generality), we are left with only the control $\varphi_S(t)$, giving

$$\partial_{\varphi_S} H_P = -\sin\varphi_S\, s_x + \cos\varphi_S\, s_y = 0 \tag{53}$$

One may note that $\dot{s}_z = \partial_{\varphi_S} H_P$, thus s_z is a constant of motion. Eq. (53) gives the condition for the regular control

$$\cos\varphi_S\, s_y = \sin\varphi_S\, s_x \tag{54a}$$

$$\implies \tan\varphi_S = \frac{s_y}{s_x} = \frac{\mathrm{Im}(s_x + is_y)}{\mathrm{Re}(s_x + is_y)} \tag{54b}$$

$$\implies \Omega_{x,y} = \frac{s_{x,y}}{s_0} \tag{54c}$$

where $s_0 = \sqrt{{s_x}^2 + {s_y}^2}$ is the modulus of the complex number $s_x + is_y$. Because both $s^2 + d^2 = {s_0}^2 + {s_z}^2 + d^2$ and s_z are constants of motion, then ${s_0}^2 + d^2$ is also a constant of motion. Furthermore, H_P can be rewritten as

$$H_P = A_\parallel d_z + s_0 = 1 \tag{55}$$

where we used $s_x = \cos\varphi_S$ and $s_y = \sin\varphi_S$.

Using these formulas for the regular control, the dynamics of $\vec{s}$ and $\vec{d}$ can be further expressed as

$$\begin{cases} \dot{s}_x = -A_\parallel d_y + \Omega_y s_z & = -A_\parallel d_y + \dfrac{s_y s_z}{s_0} \\ \dot{s}_y = A_\parallel d_x - \Omega_x s_z & = A_\parallel d_x - \dfrac{s_x s_z}{s_0} \\ \dot{s}_z = -\Omega_y s_x + \Omega_x s_y & = 0 \end{cases} \quad ; \quad \begin{cases} \dot{d}_x = -A_\parallel s_y + \Omega_y d_z & = \left(-A_\parallel + \dfrac{d_z}{s_0}\right) s_y \\ \dot{d}_y = A_\parallel s_x - \Omega_x d_z & = -\left(-A_\parallel + \dfrac{d_z}{s_0}\right) s_x \\ \dot{d}_z = -\Omega_y d_x + \Omega_x d_y & = \dfrac{1}{s_0}(s_x d_y - s_y d_x) \end{cases} \tag{56}$$

Using these formulas, we can show that $\vec{s}\cdot\vec{d}$ is a constant of motion. On another hand, using eq. (54b), one can calculate

$$\tan\varphi_S = \frac{s_y}{s_x} \implies \frac{\dot{\varphi}_s}{\cos^2\varphi_S} = \frac{\dot{s}_y s_x - s_y \dot{s}_x}{{s_x}^2} = \frac{A_\parallel(s_x d_x + s_y d_y) - \frac{s_z}{s_0}\left({s_x}^2 + {s_y}^2\right)}{{s_0}^2\cos^2\varphi_S} \tag{57a}$$

$$\implies {s_0}^2\dot{\varphi} = A_\parallel(s_x d_x + s_y d_y) - s_z s_0 = A_\parallel\, \vec{s}\cdot\vec{d} - s_z \tag{57b}$$

where we used the fact that $s_0 = 1 - A_\parallel d_z$.

Before going any further, let's do a sum up of our different constants of motions up to now.

$$\begin{cases} \vec{R_k}^2 = {R_k}^2 \\ \vec{s}^2 + \vec{d}^2 = s^2 + d^2 \\ s_z = s_z^0 \\ {s_0}^2 + d^2 = s^2 + d^2 - {s_z}^2 \\ A_\parallel d_z + s_0 = 1 \\ \vec{s} \cdot \vec{d} = p \\ {s_0}^2 \dot{\varphi}_S = C = A_\parallel\, p - s_z \end{cases} \tag{58}$$

Where we have to not forget that not all those constants are independent.

Because $s_0 = 1 - A_\parallel d_z$, we have $\dot{s}_0 = -A_\parallel \dot{d_z} = -A_\parallel (\cos\varphi_S\, d_y - \sin\varphi_S\, d_x)$, and the second derivative can be written as

$$\ddot{s}_0 = A_\parallel \dot{\varphi}_S (\cos\varphi_S\, d_x + \sin\varphi_S\, d_y) + A_\parallel \left(-A_\parallel + \frac{d_z}{s_0}\right)(\cos\varphi_S\, s_x + \sin\varphi_S\, s_y) \tag{59a}$$

$$= A_\parallel \frac{C}{{s_0}^2} \cdot \frac{1}{s_0}(s_x d_x + s_y d_y) + \left(-{A_\parallel}^2 + \frac{1 -_s 0}{s_0}\right) \cdot s_0 \left(\cos^2\varphi_S + \sin^2\varphi_S\right) \tag{59b}$$

$$= \frac{A_\parallel C}{{s_0}^3}\left(\vec{s}\cdot\vec{d} - s_z d_z\right) - {A_\parallel}^2 + 1 - s_0 \tag{59c}$$

$$= \frac{A_\parallel C}{{s_0}^3}\left(\frac{C + s_z}{A_\parallel} - s_z \frac{1 - s_0}{A_\parallel}\right) - {A_\parallel}^2 s_0 + 1 - s_0 \tag{59d}$$

$$= \frac{C^2}{{s_0}^3} + \frac{C s_z}{{s_0}^2} + 1 - (1 + {A_\parallel}^2) s_0 \tag{59e}$$

By multiplying both sides by $\dot{s}_0$ and integrating with respect to s_0, this expression can be expressed as an equivalent 2-D Hamiltonian for a particle of polar coordinate (s_0, φ_S) in a potential $U(s_0)$, with the constant ${s_0}^2 C = \dot{\varphi}$:

$$\begin{cases} E = \frac{1}{2}{\dot{s}_0}^2 + U(s_0) \\ U(X) = \frac{C^2}{2} X^{-2} + C s_z\, X^{-1} - X + \frac{1 + {A_\parallel}^2}{2} X^2 \end{cases} \tag{60}$$

Integrating the equation for s_0 gives

$${\dot{s}_0}^2 = 2\left[E - U(s_0)\right] \implies \dot{s}_0 = \epsilon\sqrt{2\left[E - U(s_0)\right]} \tag{61a}$$

$$\implies \epsilon \int_0^t d\tau = \int_0^t \frac{\dot{s}_0\, d\tau}{\sqrt{2\left[E - U(s_0)\right]}} \tag{61b}$$

$$\implies \epsilon t = \int_{s_0^0}^{s_0(t)} \frac{ds_0}{\sqrt{2\left[E - U(s_0)\right]}} \tag{61c}$$

with $\epsilon = \mathrm{sgn}(\dot{s}_0)$. We may define a new variable $v = {s_0}^{-1}$, giving $ds_0 = -v^{-2} dv$, and

$$\begin{cases} \epsilon t = \frac{1}{\sqrt{2}} \int_{s_0^0}^{s(t)} \frac{s_0\, ds_0}{\sqrt{-\frac{1 + {A_\parallel}^2}{2} {s_0}^4 + {s_0}^3 + E {s_0}^2 - C s_z s_0 - \frac{C^2}{2}}} \\ \epsilon t = -\frac{1}{\sqrt{2}} \int_{v^0}^{v(t)} \frac{dv}{v\sqrt{-\frac{C^2}{2} v^4 - C s_z v^3 + E v^2 + v - \frac{1 + {A_\parallel}^2}{2}}} \end{cases} \tag{62}$$

References

[1] Yuan H, Zeier R, Pomplun N, Glaser SJ, Khaneja N. Time-optimal polarization transfer from an electron spin to a nuclear spin. Phys Rev A. 2015 Nov;92:053414.

[2] Taminiau TH, Wagenaar JJT, van der Sar T, Jelezko F, Dobrovitski VV, Hanson R. Detection and Control of Individual Nuclear Spins Using a Weakly Coupled Electron Spin. Phys Rev Lett. 2012 Sep;109:137602. Available from: `https://link.aps.org/doi/10.1103/PhysRevLett.109.137602`.

A Preliminary results for rotating frames calculations

This part will rely on the Baker–Campbell–Hausdorff (BCH) formula:

$$\exp(aX)Y\exp(-aX) = \sum \frac{a^n}{n!}[(X)^n, Y], \tag{S1}$$

with $[(X)^n, Y] = [X, [X, ..[X, Y]]...]$ and $[(X)^0, Y] = Y$. Using the spin matrices identities $[S_i, S_j] = i\,\epsilon_{ijk}S_k$ we have:

$$[(S_z)^{(2k)}, S_{x,y}] = S_{x,y}, [(S_z)^{(2k+1)}, S_{x,y}] = \pm iS_{y,x}, \tag{S2}$$

and the Taylor serie of sine and cosine, we can show that:

$$\exp(i\omega tS_z)S_x\exp(-i\omega tS_z) = \cos(\omega t)S_x - \sin(\omega t)S_y, \tag{S3}$$

and

$$\exp(i\omega tS_z)S_y\exp(-i\omega tS_z) = \sin(\omega t)S_x + \cos(\omega t)S_y. \tag{S4}$$

We remind here the trigonometric equations:

$$\cos(a)\cos(b) = \frac{1}{2}(\cos(a+b) + \cos(a-b)), \cos(a)\sin(b) = \frac{1}{2}(\sin(a+b) - \sin(a-b)). \tag{S5}$$

For coupling terms in the form of JS_zI_z, we can use the following commutator equality:

$$[A, B] = 0 \Rightarrow [X \otimes A, Y \otimes B] = [X, Y] \otimes AB. \tag{S6}$$

Such that if we use the notation of the projector:

$$\Pi_k = |k\rangle\langle k|, \tag{S7}$$

we have:

$$[(S_zI_z)^{(2n)}, S_{x,y}] = \frac{1}{2^{2n}}S_{x,y}(\Pi_{1/2} + \Pi_{-1/2}), [(S_z)^{(2k+1)}, S_{x,y}] = \pm\frac{1}{2^{2n}}iS_{y,x}(\Pi_{-1/2} - \Pi_{1/2}). \tag{S8}$$

Thus, using the same derivation as above we can show that:

$$\exp(iJtS_zI_z)S_x\exp(-iJtS_zI_z) = \sum_{k=-1/2,1/2} \cos(kJt)S_x \otimes \Pi_k - \sin(kJt)S_y \otimes \Pi_k. \tag{S9}$$