Supplementary information

Generalized Ramsey interferometry explored with a single nuclear spin qudit

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Content

1) Ramsey sequence

We recall the expression of the evolution operator for the nuclear spin when it is under the influence of a monochromatic pulse with the pulsation ω_{pq} of a $|p\rangle \leftrightarrow |q\rangle$ Rabi oscillation with a phase shift φ:

$$\mathrm{R}_{\varphi}^{|p\rangle,|q\rangle}(\theta) = exp\left[\, i\,\theta\left(\cos\varphi\ \boldsymbol{\sigma}_x^{|p\rangle,|q\rangle} + \sin\varphi\ \boldsymbol{\sigma}_y^{|p\rangle,|q\rangle}\right)/2\right] \quad (1)$$

Given a state $|p\rangle$, the two paths of an interferometer are built by creating a quantum superposition with the partner state $|q\rangle$ of a $|p\rangle \leftrightarrow |q\rangle$ Rabi oscillation. This is merely achieved by applying a π/2 pulse of pulsation ω_{pq} during the time $\tau = (\pi/2)/\omega_{pq}$:

$$|p\rangle \ \rightarrow\ \mathrm{R}_{\varphi}^{|p\rangle,|q\rangle}(\pi/2)|p\rangle = \frac{|p\rangle - i\, e^{i\varphi}\,|q\rangle}{\sqrt{2}} \qquad (2)$$

The states $|p\rangle$ and $|q\rangle$ can be manipulated separately by microwave pulses of distinct pulsations what will feed them with independent phases φ_p and φ_q:

$$\frac{|p\rangle - i\,e^{i\varphi}\,|q\rangle}{\sqrt{2}} \rightarrow \frac{e^{i\varphi_p}\,|p\rangle - i\,e^{i\varphi}\,e^{i\varphi_q}|q\rangle}{\sqrt{2}} \qquad (3)$$

At any instant of a manipulation the signals of the two arms of the interferometer can be merged back *via* the application of the same π/2 pulse:

$$\mathrm{R}_{\varphi}^{|p\rangle,|q\rangle}(\pi/2)\frac{e^{i\varphi_p}\,|p\rangle - i\,e^{i\varphi}\,e^{i\varphi_q}|q\rangle}{\sqrt{2}} = \frac{(e^{i\varphi_p} - e^{i\varphi_q})\,|p\rangle - i\,e^{i(\varphi-\varphi_p-\varphi_q)}\,(e^{-i\varphi_p} - e^{-i\varphi_q})|q\rangle}{2} \qquad (4)$$

In this final state the probability of being in the state $|p\rangle$ and the one of being in the state $|q\rangle$ are respectively given by

$$\cos^2\left(\frac{\varphi_p-\varphi_q}{2}\right) \quad \text{and} \quad \sin^2\left(\frac{\varphi_p-\varphi_q}{2}\right) \qquad (5)$$

which reveals the difference in the phases separately accumulated by the two states from the time at which the first π/2 pulse was applied up to the time at which the second π/2 pulse was started.

2) Geometric phase

Considering a nuclear spin $|I\rangle$ evolving, with a Hamiltonian H, in its Hilbert space of dimensions 2I+1 and their associated fiber bundle of dimension one. The phase of a spin state, defined as the argument of its inner product, is affected both by the evolution in the Hilbert space (dynamic phase) and by the continuous connection in between each fiber "traveled" by the spin during the dynamic (geometric phase).

As a consequence, when a spin describes a closed path in its Hilbert space, depending on the geometry of this path along the fibers, the initial and the final state can differ from a geometric phase factor. The expression of this phase accumulated on a circuit C is[30]:

$$\gamma_G(C) = i\oint_C \left\langle I(R)\middle|\frac{\partial}{\partial R}I(R)\right\rangle . dR \qquad (6)$$

To create spin trajectory, it is convenient to use rotation on its associated Hilbert space. We derive the general expression of the inner product in the integral considering first a rotation of angle θ of generator I_y followed by a rotation of angle φ of generator I_z:

$$\left\langle I(R)\middle|\frac{\partial}{\partial R}I(R)\right\rangle = \left\langle I\middle|\exp\left(i\theta I_y\right)\exp(i\varphi I_z)\frac{\partial}{\partial\theta}\exp(-i\varphi I_z)\exp\left(-i\theta I_y\right)\middle|I\right\rangle.\partial\theta + \left\langle I\middle|\exp\left(i\theta I_y\right)\exp(i\varphi I_z)\frac{\partial}{\sin\theta\partial\varphi}\exp(-i\varphi I_z)\exp\left(-i\theta I_y\right)\middle|I\right\rangle.\sin\theta\,\partial\varphi = \langle I|-\mathrm{i}I_y|I\rangle.\partial\theta + \left\langle I\middle|\exp\left(i\theta I_y\right)\frac{-iJ_z}{\sin\theta}\exp\left(-i\theta I_y\right)\middle|I\right\rangle.\sin\theta\,\partial\varphi \quad (7)$$

Because I_y is off-diagonal, the first term is equal to zero. Using commutation relation, we obtain finally the expression:

$$\left\langle I(R)\middle|\frac{\partial}{\partial R}I(R)\right\rangle = \left\langle I\middle|\exp\left(i\theta I_y\right)\frac{-iI_z}{\sin\theta}\exp\left(-i\theta I_y\right)\middle|I\right\rangle.\sin\theta\,\partial\varphi = \left\langle I\middle|\frac{-i\left(J_z\cos\theta+J_x\sin\theta\right)}{\sin\theta}\middle|I\right\rangle.\sin\theta\,\partial\varphi = -i\,I\cos\theta\;\mathrm{d}\varphi \quad (8)$$

In all the protocol we present, the circuit C is the following:

$$\oint_C = \int_{\theta=\pi}^{0}\int_{\varphi=0}^{\varphi'}\int_{\theta=0}^{\pi} \quad (9)$$

giving the final expression of the geometric phase:

$$\gamma_G = 2I\varphi \quad (10)$$

When we drive only one transition, it is equivalent to measure the phase accumulated by a spin ½, giving the expression $\gamma_G = \varphi$. When we drive simultaneously two transitions, it is equivalent to measure the phase accumulated by a spin 1, giving the expression $\gamma_G = 2\varphi$.

3) iSWAP quantum gate

We display the comparison in between the theoretical and the experimental evolution of the different state populations as the function of the pulse duration.

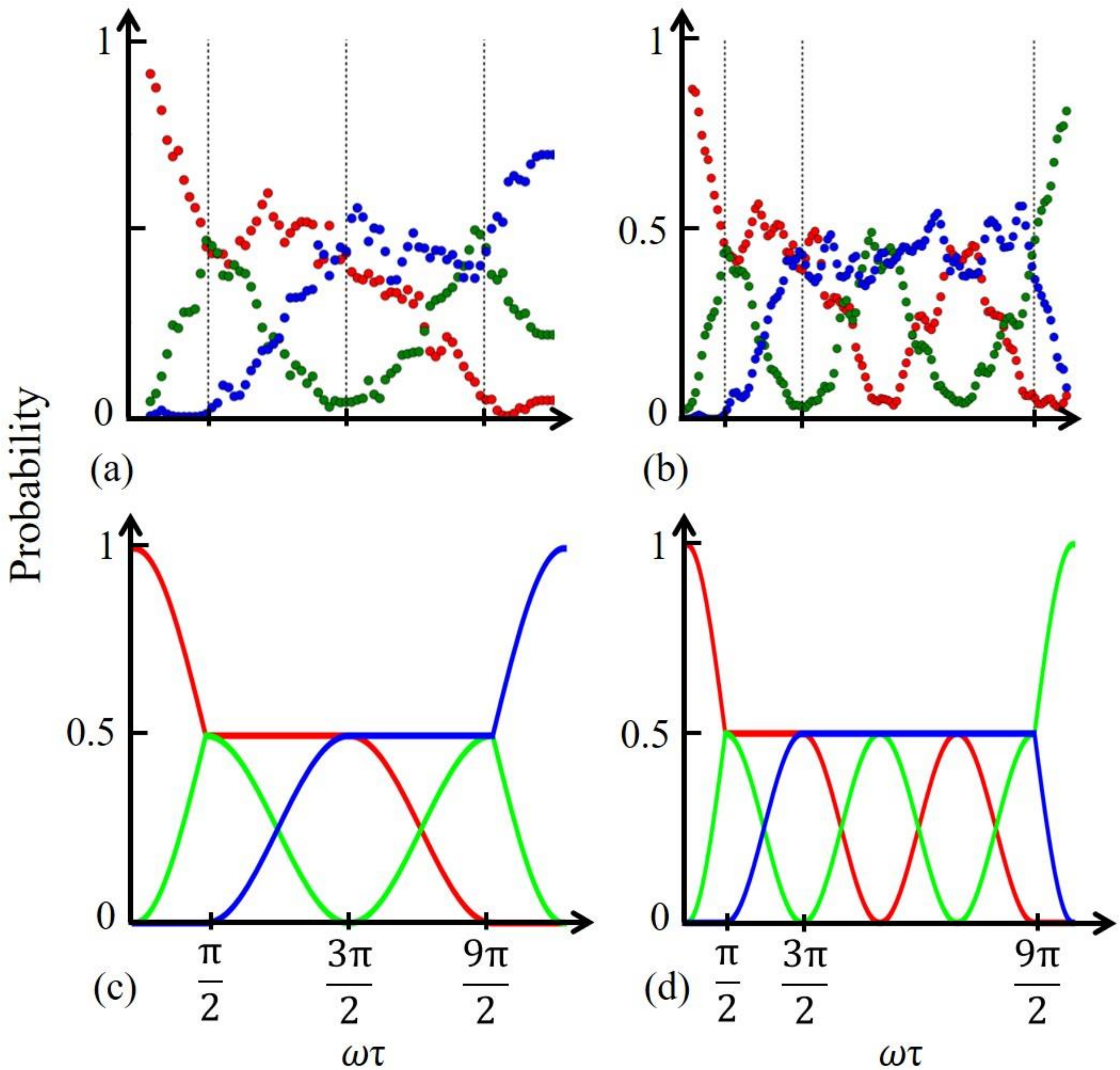

Figure. **1SM**. Experimental (a-b) and theoretical (c-d) evolution of each state population as the function of the pulse duration for the two iSWAP phase measurement.

4) 3-state coherence time measurement

A complete derivation of the interaction of a multi-level system with a multi-frequency pulse is detailed in supplementary material of Ref. 12. For a 3-state (of respective eigen-energy ε_0, ε_1 and ε_2) 2-frequency pulse, associated with Rabi pulsation ω_1 and ω_2 between the state 0 and 1 and states 1 and 2 respectively. The expression of the Hamiltonian in the generalized rotating frame is the following:

$$H_{g.r.f} = \hbar \begin{pmatrix} 0 & \frac{\omega_1}{2} & 0 \\ \frac{\omega_1}{2} & \delta_1 & \frac{\omega_2}{2} \\ 0 & \frac{\omega_2}{2} & \delta_2 \end{pmatrix} \qquad (11)$$

with $\delta_1 = (\varepsilon_1 - \varepsilon_0)/\hbar - \omega_1$ and $\delta_2 = (\varepsilon_2 - \varepsilon_1)/\hbar - \omega_2$. In our case, to simplify the dynamic, we decide to drive both resonance such that $\omega_1 = \omega_2 = \omega$ and that $\delta_2 = 0$, leading to the Hamiltonian:

$$H = \frac{\hbar}{2} \begin{pmatrix} 0 & \omega & 0 \\ \omega & 2\delta & \omega \\ 0 & \omega & 0 \end{pmatrix} \qquad (12)$$

For a d-level system, a Hadamard gate creates a coherent superposition of all the states of the system:

$$U_{Had}|\Psi_i\rangle = \frac{1}{\sqrt{d}} \sum_{n=0}^{d-1} |n\rangle = |\Psi_s\rangle \qquad (13)$$

An efficient way of implementing this gate is to create a resonant condition in between the initial state and the superposed state:

$$\langle \Psi_s | H | \Psi_s \rangle = \langle \Psi_i | H | \Psi_i \rangle \qquad (14)$$

After derivation, choosing $|-1/2\rangle$ as an initial state, this condition fixes the relation in between δ and ω: $\omega = \delta$

Because of the resonant condition, the full system oscillates in between the initial state and the superposed state. As a consequence, starting from state $|-1/2\rangle$, after a half period (T/2= $\frac{\pi\sqrt{3}}{3\Omega}$), the system is fully in the superposed state:

$$U_{Had}|\Psi_i\rangle = \exp\left(-iH\frac{\pi\sqrt{3}}{3\omega}\right).\begin{pmatrix}0\\1\\0\end{pmatrix} = \frac{1}{\sqrt{3}}\begin{pmatrix}1\\1\\1\end{pmatrix} \qquad (15)$$

Then the system is let under free evolution, meaning that the Hamiltonian is:

$$H_W = \frac{\hbar}{2}\begin{pmatrix}0 & 0 & 0\\0 & 2\omega & 0\\0 & 0 & 0\end{pmatrix} \qquad (16)$$

During a time τ:

$$U_W.U_{Had}|\Psi_i\rangle\frac{1}{\sqrt{3}}\exp(-iH_W\tau).\begin{pmatrix}1\\1\\1\end{pmatrix} = \frac{1}{\sqrt{3}}\begin{pmatrix}1\\e^{i\omega\tau}\\1\end{pmatrix} \qquad (17)$$

Finally the second Hadamard gate is implemented, which results in the state:

$$U_{Had}.U_W.U_{Had}|\Psi_i\rangle = \frac{1}{\sqrt{3}}\exp\left(-iH\frac{\pi\sqrt{3}}{3\omega}\right).\begin{pmatrix}1\\e^{i\omega\tau}\\1\end{pmatrix} = \frac{1}{3}\begin{pmatrix}e^{i\omega\tau}-1\\e^{i\omega\tau}+2\\e^{i\omega\tau}-1\end{pmatrix} \qquad (18)$$