

# Supplementary Information

## A.1 - Brief introduction to quantum systems

There are several formalisms in which quantum mechanics can be expressed, including the one making use of Hilbert spaces and density matrices [3], which we shall denote as *Hilbert* for clarity. In the *Hilbert* formalism, the theory is expressed in terms of states,  $|\Psi\rangle$ , and transitions between them.

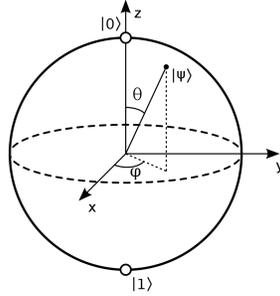


Figure 1: The set of points definable on the surface of the Bloch sphere, compose the state space of a *Hilbert* space of dimension 2 (*qubit*) of basis  $|0\rangle$ ,  $|1\rangle$ , i.e.  $|\Psi\rangle = \lambda|0\rangle + \beta|1\rangle$  with  $|\lambda|^2 + |\beta|^2 = 1$ .

The states may be expanded on an *eigenbasis* of the Hilbert space,

$$|\Psi\rangle = \sum_i \lambda_i |\Psi_i\rangle, \text{ with } \lambda_i \in \mathbb{C}$$

where  $\sum_i |\lambda_i|^2 = 1$ . The *eigenbasis* states are often taken as the *eigenstates* of a Hamiltonian and known in quantum mechanics as the *stationary states*, while  $|\Psi\rangle$  is an example of the so-called *superposition states*.

Furthermore, quantum systems can also be composed and form new systems through the *Kronecker* product of the Hilbert spaces, corresponding to two sub-systems (in this case 1 and 2):

$$\mathcal{H}_1 \otimes \mathcal{H}_2 \tag{1}$$

The analysis of this operator quickly leads to another very relevant property of quantum mechanics, which is the existence of *non-separable* states, i.e. states that cannot be written as a Cartesian separable product of the subsystems 1 and 2, as for instance happens in the so-called Bell states,

$$|\Psi\rangle = \frac{1}{\sqrt{2}} (|0_1 0_2\rangle + |1_1 1_2\rangle), \tag{2}$$

which are a possible state of the  $\otimes$  on two qubits. This phenomenon is the so-called *entanglement*, a vital component of many quantum technologies such as quantum information. An extension of *pure* states represented by vectors in

the Hilbert space leads to the density matrices, which can also describe the so called *mixed* states. A mixed state can be considered as an mixture of several pure states, with certain statistical weights, therefore it cannot be described by a vector in the Hilbert space. A (Hermitian) density matrix (or operator) can be written as

$$\hat{\rho} = \sum_{\alpha} P_{\alpha} |\Phi_{\alpha}\rangle \langle \Phi_{\alpha}|$$

where  $P_{\alpha}$  are real numbers between 0 and 1 and  $|\Phi_{\alpha}\rangle \langle \Phi_{\alpha}|$  is known as the *projection* operator of a state  $\Phi_{\alpha}$ . For a *pure* state all  $P_{\alpha} = 0$  except for one, which is equal to unity. By expanding  $\Phi_{\alpha}$  on a basis of stationary states, one can obtain the matrix representation of  $\hat{\rho}$  in that basis. The density matrix of a system composed of two parts,  $A$  and  $B$ , allows for a simple criterion for checking whether these parts are entangled or not in a particular state,  $\hat{\rho}_{A+B}$  (see, e.g. [1]). To distinguish between entangled and separable states, one needs to calculate the density matrix of subsystem  $A$  (or  $B$ ) as the partial trace of  $\hat{\rho}_{A+B}$  and check the trace of  $\hat{\rho}_A^2$ . If it is equal to unity, the subsystem  $A$  is in a pure state and the whole system is separable.

While the Hamiltonian formalism of quantum mechanics permits properly describe *closed* systems (and, in principle, universe as a whole), in practice, for instance in quantum technology, one deals with systems that are not closed, i.e. there is a free uncontrollable element, not necessarily conservative, interacting the system under study. The theory that studies these kind of systems is a branch of quantum mechanics known as *open quantum systems* [2]. In fact, an open system is nothing more than a *closed system* composed of two subsystems, the *system* and the *environment*, in which the system plus environment Hilbert space is expressed as  $\mathcal{H}_{S+E}$ .

The overall effect of the environment in the system can be obtained by a *tracing* operation over the environment Hilbert space:

$$\mathcal{H}_S = Tr_E[\mathcal{H}_{S+E}]. \quad (3)$$

## A.2 - Solution of the Schrödinger and Liouville equations for the two-molecule system

Making use of the Schrödinger equation ( $\hbar = 1$ ):

$$i \frac{\delta}{\delta t} |\Psi\rangle = \hat{H} |\Psi\rangle \quad (4)$$

and writing the state  $|\Psi\rangle$  in the site basis as:

$$|\Psi\rangle = \alpha |0\rangle + \beta |1\rangle \quad (5)$$

where  $|\alpha|^2 + |\beta|^2 = 1$ . One gets by applying the Schrödinger equation to the state  $|\Psi\rangle$  and multiplying it by  $\langle 0|$  and  $\langle 1|$ :

$$i\dot{\alpha} = \epsilon_0\alpha + J\beta \quad (6)$$

$$i\dot{\beta} = \epsilon_1\beta + J\alpha \quad (7)$$

By making the substitutions  $\alpha = ae^{-i\epsilon_0 t}$  and  $\beta = be^{-i\epsilon_1 t}$  so that:

$$\dot{\alpha} = \dot{a}e^{-i\epsilon_0 t} - i\epsilon_0\alpha \quad (8)$$

$$\dot{\beta} = \dot{b}e^{-i\epsilon_1 t} - i\epsilon_1\beta \quad (9)$$

one has, taking into account the substitutions and Eqs. (6) and (7):

$$i\dot{a} = Je^{-iwt}b \quad (10)$$

$$i\dot{b} = Je^{iwt}a \quad (11)$$

where  $w = \epsilon_1 - \epsilon_0$ . Then the variable  $c$  is introduced as  $c = ae^{iwt}$ . One can observe through Eq. (10) that  $\dot{a} = (\dot{c} - iwc)e^{-iwt}$  so that  $i(\dot{c} - iwc) = Jb$ . Differentiating with respect to time and using Eq. (11), one has:

$$\ddot{c} - iw\dot{c} + J^2c = 0 \quad (12)$$

By seeking solutions as  $c = e^{i\lambda t}$ :

$$-\lambda^2 + w\lambda + J^2 = 0 \quad (13)$$

one gets:

$$\lambda_{0,1} = \frac{w}{2} \pm \sqrt{\frac{w^2}{4} + J^2} = \frac{1}{2}(w \pm \Omega) \quad (14)$$

where  $\Omega = \sqrt{w^2 + 4J^2}$ . The solution of Eq. (12) is:

$$c = Ae^{i\lambda_0 t} + Be^{i\lambda_1 t} \quad (15)$$

then,

$$a(t) = Ae^{i\frac{\Omega-w}{2}t} + Be^{-i\frac{\Omega+w}{2}t} \quad (16)$$

and  $b(t)$  is found by using equation (11):

$$b(t) = \frac{\Omega + w}{2J}Be^{i\frac{w-\Omega}{2}t} - \frac{\Omega - w}{2J}Ae^{i\frac{w+\Omega}{2}t} \quad (17)$$

The constants  $A$  and  $B$  depend on the initial conditions. If the donor molecule is the only one excited ( $|0\rangle$ ) at the initial time  $t = 0$ , then  $\alpha(0) = 1$  and  $\beta(0) = 0$ . Thus,  $a(0) = 1$  and  $b(0) = 0$  and one gets  $A = \frac{\Omega+w}{2\Omega}$  and  $B = \frac{\Omega-w}{2\Omega}$ . The solutions are:

$$a(t) = \frac{\lambda_0}{\Omega}e^{-i\lambda_0 t} - \frac{\lambda_1}{\Omega}e^{-i\lambda_1 t} \quad (18)$$

$$b(t) = \frac{J}{\Omega}[e^{i\lambda_0 t} - e^{i\lambda_1 t}] \quad (19)$$

and the system's density matrix becomes:

$$\rho = |\Psi\rangle\langle\Psi| = \begin{pmatrix} |\alpha|^2 & \alpha\beta^\dagger \\ \alpha^\dagger\beta & |\beta|^2 \end{pmatrix} = \begin{pmatrix} |a(t)|^2 & a(t)b^\dagger(t)e^{iwt} \\ a^\dagger(t)b(t)e^{-iwt} & |b(t)|^2 \end{pmatrix}. \quad (20)$$

The time dependence of the non-diagonal elements of the density matrix in the energy basis can be obtained much easier. Consider the quantum master Liouville equation applied to a closed system  $\rho$  subjected to a Hamiltonian  $H$ , defined as:

$$\frac{d\rho}{dt} = -i[H, \rho] \quad (21)$$

Writing the density matrix in the energy eigenbasis (where the Hamiltonian is diagonal) yields:

$$\frac{d\rho_{ij}}{dt} = -i(E_i - E_j)\rho_{ij}, \quad (22)$$

where  $E_i$  is the energy eigenvalue of the  $i$ th energy eigenstate. The populations (matrix elements  $\rho_{ij}$ , where  $i = j$ ) remain constant and the coherence terms (matrix elements  $\rho_{ij}$ , where  $i \neq j$ ) evolve oscillating in time as:

$$\rho_{ij}(t) = e^{-i(E_i - E_j)t} \rho_{ij}(0) \quad (23)$$

For the two molecule system, the Hamiltonian

$$\hat{H}_S = \begin{pmatrix} \epsilon_0 & J \\ J & \epsilon_1 \end{pmatrix}. \quad (24)$$

the energy eigenvalues  $E_0$  and  $E_1$  become, upon diagonalization, as

$$E_0 = \frac{1}{2}[\epsilon_0 + \epsilon_1 + \sqrt{(\epsilon_0 - \epsilon_1)^2 + 4J^2}] \quad (25)$$

and

$$E_1 = \frac{1}{2}[\epsilon_0 + \epsilon_1 - \sqrt{(\epsilon_0 - \epsilon_1)^2 + 4J^2}]. \quad (26)$$

Therefore, the coherence terms in Eq. (23) evolve as:

$$\rho_{ij}(t) = e^{-i\omega t} \rho_{ij}(0) \quad (27)$$

where the frequency of the quantum beating is given by  $\Omega = \sqrt{(\epsilon_0 - \epsilon_1)^2 + 4J^2}$ . This corresponds to the equation,

$$\rho_{ij}(t) = e^{-it\sqrt{(\epsilon_0 - \epsilon_1)^2 + 4J^2}/\hbar} \rho_{ij}(0), \quad i \neq j \quad (28)$$

,presented in the main text. The period of the oscillations is  $\frac{2\pi}{\Omega}$ .

### A.3 - Circuits for the coherent regime

This annex contains the optimized circuits used to build the quantum simulations of section *No decoherence regime* for the near resonance (figure 2) and off resonance (figure 3) regimes.

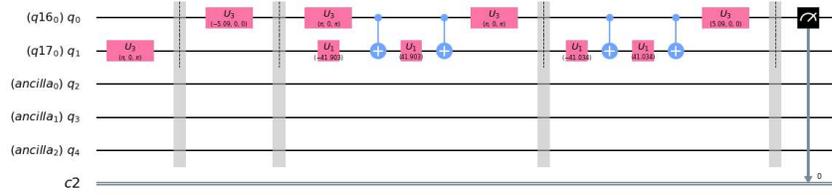


Figure 2: Optimized quantum circuit for the near-resonance system simulation.

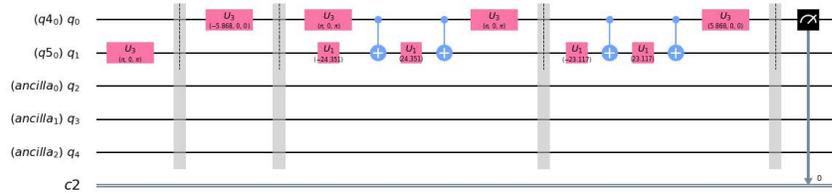


Figure 3: Optimized quantum circuit for the non-resonance system simulation.

## References

- [1] Stephen Barnett. *Quantum information*, volume 16. Oxford University Press, 2009.
- [2] Heinz-Peter Breuer, Francesco Petruccione, et al. *The theory of open quantum systems*. Oxford University Press on Demand, 2002.
- [3] Michael A Nielsen and Isaac Chuang. *Quantum computation and quantum information*. Cambridge University Press, 2010.