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Realization of controllable open system with NMR

To cite this article: Le Bin Ho *et al* 2019 *New J. Phys.* **21** 093008

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PAPER

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RECEIVED
13 March 2019REVISED
12 July 2019ACCEPTED FOR PUBLICATION
5 August 2019PUBLISHED
4 September 2019

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Abstract

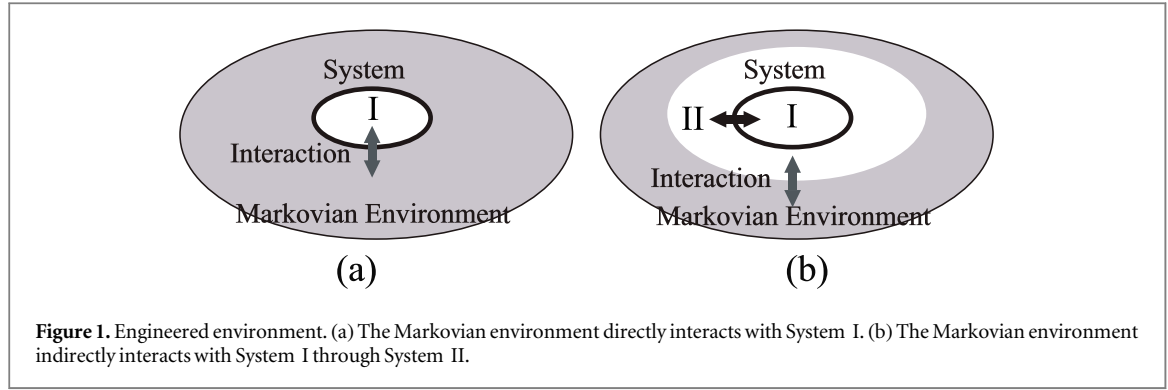
An open quantum system is now attracting much attention because a quantum device such as quantum computers and quantum sensors is an emerging technology. Here, we present a model of the open system that shows either time-homogeneous Markovian relaxations or non-Markovian relaxations depending on its parameters that we can control. This model is fully described with the master equation that is analytically solvable. More importantly, this model can be easily realized with molecules in isotropic liquids and measured with NMR techniques.

1. Introduction

It is important to understand open systems both in science and technology because a quantum device such as quantum computers and quantum sensors is an emerging technology: these devices are inevitably quantum open systems however one tries to isolate them from environment. For not only theoretical interest but also practical one, understanding of characteristics of dynamics of these open systems (time-homogeneous Markovian, time-inhomogeneous Markovian or non-Markovian [1, 2]) is important in order to protect them from their environments [3–5]: it is theoretically pointed out [6, 7] that an entangled sensor can beat the standard quantum limit (SQL) with time-inhomogeneous Markovian noise while it usually shows the same scaling as the SQL with Markovian noise. The SQL is the consequence of the central-limit theorem [8] and is considered as a classical scaling. We note, however, that time-homogeneous Markovian dynamics let quantum sensors achieve a sub SQL scaling in particular situations, see [2] for more details.

Both experimental and theoretical efforts to understand open systems have been carried out. There are the experiments with ultra cold atoms [9], ions in traps [10], optics [11], and cold electric circuits [12] to study open systems. Experimental controls of the degree of non-Markovianity in optics [13–15], ion traps [16], NV centers in diamond [17–19], and NMR [20] are most important for our work. We have also reported some preliminary results [21, 22]. Among theoretical works, the pseudomode approach to an open-system such as [23, 24] and the extended collision model [25] are important for our work.

We have been realizing an engineered environment and studying methods protecting a system from environment [21, 26, 27]. Our engineered environment may be regarded as a realization of the pseudomode [23, 24] or the extended collision model [25]. So far we could only numerically solve the dynamics of a system in the engineered environment. In this work, we provide analytical solutions of the systems in the various engineered environments and compare the solutions with experimental observations in details. The systems show either time-homogeneous Markovian, time-inhomogeneous Markovian, or non-Markovian relaxations depending on their parameters. Our model is fully described with the Lindblad master equation [28] that is analytically solvable [29–31]. Since a simpler model environment can be equivalent to a more complicated environment in terms of a dynamics of an open system [24], our experimental realization of an open system must be useful to understand the open system.



The rest of the paper is organized as follows. In section 2, we present a theoretical model to realize an open system that show either time-homogeneous Markovian relaxations or non-Markovian relaxations depending on its parameters, and also derive its analytical solution. Then we present its experimental realization using star-topology molecules in isotropic liquids in section 3. The last section 4 is devoted to conclusion. In section appendix, detailed calculations are shown.

2. Engineered environment

First of all, in order to avoid any confusions, we define the Markovianity and non-Markovianity as follows [1, 2]. If the information always flows from a system (that we are interested in) to an environment regardless its flow rate, we call the environment Markovian. When its flow rate is constant, then it is a time-homogeneous Markovian environment. On the other hand, when its flow rate is time dependent, then it is a time-inhomogeneous one. If the information flows back to the system from the environment at some moments, then it is regarded as a non-Markovian environment. Note that, although non-exponential decay was occasionally considered as a non-Markovian property in some past literatures [6], we do not adopt such a definition in our paper. Non-Markovianity is more rigorously measured by using a trace distance [32] or with the CP-divisibility [33].

We illustrate our idea to obtain various open systems or to engineer the environment in figure 1 [21, 22, 26, 27], see also [23, 24]. If a well-defined quantum system (such as a qubit) shows an exponential relaxation, the system interacts with a surrounding short-time memory (or, Markovian) environment. On the other hand, if its relaxation is non-exponential, the surrounding one should have a long-time memory [34]. We realize such a long-time memory environment in two steps: a well-defined primary system (hereafter called System I) interacts only with an ancillary system (hereafter called System II) and System II is under the influence of a Markovian environment. Note that System I and II are parts of one composite system in our idea. We regarded a proper kind of molecule in isotropic liquid as this composite system in our experiments discussed in section 3. Our model may be considered as an abstraction of systems that show non-exponential relaxations [16–19].

In our previous works [21, 27], we employed numerical simulations with the idea of quantum channels in order to evaluate the above models. In this paper, we will start from the Lindblad master equation [28] and solve it analytically [29–31]. The Lindblad master equation is generally given as

$$\frac{d\rho}{dt} = -i[H, \rho] + \sum_{k,j} c_{k,j} \left(\mathcal{L}_k \rho \mathcal{L}_j^\dagger - \frac{1}{2} \{ \mathcal{L}_k^\dagger \mathcal{L}_j, \rho \} \right), \quad (1)$$

where H is the system Hamiltonian, $(c_{k,j})$ is a positive definite matrix, \mathcal{L}_j are the Lindblad operators to represent the influence of an environment on the system, and $[*, *], \{*, *\}$ denotes an (anti-) commutator. We take the natural unit system where $\hbar = 1$ in this paper.

We introduce a Markovian environment that induces random flip-flop transitions of the qubit. In this case, the Lindblad master equation is given as

$$\frac{d\rho}{dt} = -i[H, \rho] + \sum_{\pm} \gamma \left(2 \frac{\sigma_{\mp} \rho \sigma_{\pm}}{4} - \left\{ \frac{\sigma_{\pm} \sigma_{\mp}}{4}, \rho \right\} \right), \quad (2)$$

where $\sigma_{\pm} = \frac{\sigma_x \pm i\sigma_y}{2}$, σ_k ($k = x, y, z$) is a standard Pauli matrix acting on the qubit, and γ is the flip-flopping rate. This can be also interpreted as an amplitude damping channel [35].

2.1. Markovian environment

Let us consider the simplest case where System I is a qubit (a nuclear spin in a magnetic field in section 3) and interacts directly with the Markovian environment, as shown in figure 1(a). Equation (2) becomes

$$\frac{d\rho_0}{dt} = -i\left[\omega\frac{\sigma_z}{2}, \rho_0\right] + \sum_{\pm} \gamma_0 \left(2\frac{\sigma_{\mp}\rho_0\sigma_{\pm}}{4} - \left\{ \frac{\sigma_{\pm}\sigma_{\mp}}{4}, \rho_0 \right\} \right), \quad (3)$$

where γ_0 is the flip-flopping rate of System I caused by the Markovian environment. Because a nuclear spin acting as System I in section 3 will be considered in a rotating frame of which frequency is the same as the Larmor frequency of the spin, we will set $\omega = 0$ in equation (3). This differential equation is easily solved by using a Bloch vector representation of the density matrix $\rho_0 = x\frac{\sigma_x}{2} + y\frac{\sigma_y}{2} + z\frac{\sigma_z}{2} + \frac{\sigma_0}{2}$, where σ_0 is the identity matrix of dimension 2. Equation (3) becomes

$$\frac{d}{dt} \begin{pmatrix} 1+z & x-iy \\ x+iy & 1-z \end{pmatrix} = -\frac{\gamma_0}{2} \begin{pmatrix} 2z & x-iy \\ x+iy & -2z \end{pmatrix},$$

or

$$\frac{dw(t)}{dt} = -\frac{\gamma_0}{2}w(t), \text{ and } \frac{dz(t)}{dt} = -\gamma_0z(t), \quad (4)$$

where $w(t) = x(t) + iy(t)$. Then, we obtain

$$w(t) = w(0)e^{-\gamma_0 t/2}, \text{ and } z(t) = z(0)e^{-\gamma_0 t}.$$

Thus, System I shows a time-homogeneous, or exponential, relaxation under the influence of Markovian environment.

2.2. Non-Markovian environment with a single qubit in System II

Let us consider the case shown in figure 1(b) where both System I and II consist of a single qubit. We call this a $(1 + 1)$ system. These qubits are realized with hetero-nuclear spins in one molecule in section 3 and they are considered in the individual rotating frame. Therefore, the system Hamiltonian only consists of the interaction between them. We assume this Hamiltonian as in the following form.

$$H_I = J \frac{\sigma_z \otimes \sigma_z}{4},$$

where J represents a strength of the interaction. This is a typical interaction between hetero-nuclear spins in one molecule under the secular approximation [36]. Note also that this interaction can only cause pure dephasing [2] on System I through System II. This interaction is essential for our idea and allows the Markovian environment to influence System I through System II. For simplicity, we assume that there is no direct influence of the Markovian environment on System I. The master equation is, then, given as

$$\frac{d\rho_1}{dt} = -i[H_I, \rho_1] + \sum_{\pm} \gamma (2\mathcal{L}_{\mp}^{(1)}\rho_1\mathcal{L}_{\pm}^{(1)} - \{\mathcal{L}_{\pm}^{(1)}\mathcal{L}_{\mp}^{(1)}, \rho_1\}), \quad (5)$$

where $\mathcal{L}_k^{(1)} = \sigma_0 \otimes \sigma_k/2$, ($k = +, -$). We assign the first (second) qubit to System I (II). This master equation can be interpreted to realize the extended collision model of relaxation [25], as we discussed in [22]. The density matrix of the total system is given as

$$\begin{aligned} \rho_1 &= \left(x_{\uparrow}\frac{\sigma_x}{2} + y_{\uparrow}\frac{\sigma_y}{2} + z_{\uparrow}\frac{\sigma_z}{2} + \frac{\sigma_0}{2} \right) \otimes \frac{|\uparrow\rangle\langle\uparrow|}{2} \\ &+ \left(x_{\downarrow}\frac{\sigma_x}{2} + y_{\downarrow}\frac{\sigma_y}{2} + z_{\downarrow}\frac{\sigma_z}{2} + \frac{\sigma_0}{2} \right) \otimes \frac{|\downarrow\rangle\langle\downarrow|}{2} = \frac{1}{4} \begin{pmatrix} 1+z_{\uparrow} & 0 & w_{\uparrow}^* & 0 \\ 0 & 1+z_{\downarrow} & 0 & w_{\downarrow}^* \\ w_{\uparrow} & 0 & 1-z_{\uparrow} & 0 \\ 0 & w_{\downarrow} & 0 & 1-z_{\downarrow} \end{pmatrix}, \end{aligned} \quad (6)$$

where $w_k = x_k + iy_k$ ($k = \uparrow, \downarrow$). We take this initial state because we intend to study the case when only System I deviates from the maximally mixed state at the $t = 0$ moment. Then, substituting ρ_1 into the right hand side of equation (5) gives

$$\frac{1}{8} \begin{pmatrix} 0 & 0 & -(\gamma - iJ)w_{\uparrow} + \gamma w_{\downarrow}^* & 0 \\ 0 & 0 & 0 & (\gamma w_{\uparrow} - (\gamma + iJ)w_{\downarrow})^* \\ -(\gamma - iJ)w_{\uparrow} + \gamma w_{\downarrow} & 0 & 0 & 0 \\ 0 & \gamma w_{\uparrow} - (\gamma + iJ)w_{\downarrow} & 0 & 0 \end{pmatrix}.$$

The master equation (5) is simplified to the following simultaneous differential equations,

$$\begin{aligned}\frac{dw_{\uparrow}}{dt} &= -\frac{\gamma - iJ}{2}w_{\uparrow} + \frac{\gamma}{2}w_{\downarrow}, \quad \frac{dz_{\uparrow}}{dt} = 0, \\ \frac{dw_{\downarrow}}{dt} &= \frac{\gamma}{2}w_{\uparrow} - \frac{\gamma + iJ}{2}w_{\downarrow}, \quad \text{and} \quad \frac{dz_{\downarrow}}{dt} = 0.\end{aligned}\quad (7)$$

Note that there is no time evolution of the z -component of the density matrix because $H_1 \propto \sigma_z \otimes \sigma_z$.

Equation (7) can be solved as follows. It is compactly presented as

$$\frac{d\vec{w}_1}{dt} = M\vec{w}_1 \quad \text{and} \quad \frac{d\vec{z}_1}{dt} = 0, \quad (8)$$

where $\vec{w}_1 = \begin{pmatrix} w_{\uparrow} \\ w_{\downarrow} \end{pmatrix}$, $\vec{z}_1 = \begin{pmatrix} z_{\uparrow} \\ z_{\downarrow} \end{pmatrix}$ and

$$M = \frac{1}{2} \begin{pmatrix} -\gamma + iJ & \gamma \\ \gamma & -\gamma - iJ \end{pmatrix} = -\frac{\gamma}{2}\sigma_0 + \frac{\gamma}{2}\sigma_x + \frac{iJ}{2}\sigma_z. \quad (9)$$

M can be diagonalized with V ,

$$V = \begin{pmatrix} \frac{-\sqrt{\gamma^2 - J^2} + iJ}{\gamma} & 1 \\ \frac{\sqrt{\gamma^2 - J^2} + iJ}{\gamma} & 1 \end{pmatrix},$$

such that

$$\Lambda = VMV^{-1} = \begin{pmatrix} \frac{-\gamma - \sqrt{-J^2 + \gamma^2}}{2} & 0 \\ 0 & \frac{-\gamma + \sqrt{-J^2 + \gamma^2}}{2} \end{pmatrix}.$$

For example, the solution with the initial values of $w_{\uparrow}(0) = w_{\downarrow}(0) = 1$ are given as,

$$\begin{aligned}w_{\uparrow}(t) &= e^{-\gamma t/2} \left(\cosh \frac{t\sqrt{-J^2 + \gamma^2}}{2} + \frac{iJ + \gamma}{\sqrt{-J^2 + \gamma^2}} \sinh \frac{t\sqrt{-J^2 + \gamma^2}}{2} \right), \\ w_{\downarrow}(t) &= e^{-\gamma t/2} \left(\cosh \frac{t\sqrt{-J^2 + \gamma^2}}{2} + \frac{-iJ + \gamma}{\sqrt{-J^2 + \gamma^2}} \sinh \frac{t\sqrt{-J^2 + \gamma^2}}{2} \right).\end{aligned}$$

Note that $z_{\uparrow} = z_{\downarrow} = 0$ in this case. When System II is traced out, the dynamics of System I, $w(t)$, is given as,

$$w(t) \equiv \frac{w_{\uparrow}(t) + w_{\downarrow}(t)}{2} = e^{-\gamma t/2} \left(\cosh \frac{t\sqrt{-J^2 + \gamma^2}}{2} + \frac{\gamma}{\sqrt{-J^2 + \gamma^2}} \sinh \frac{t\sqrt{-J^2 + \gamma^2}}{2} \right). \quad (10)$$

As we increase γ/J , the relaxation becomes slower and the behavior becomes closer to an exponential decay. This may be understood in terms of the motional narrowing [36]. When System II flip-flops much more frequently than the time scale determined by $1/J$, System I has no time to accumulate a significant phase changes between the flip-flops. Since our time scale is $1/J$ and the average times of $|\uparrow\rangle$ and $|\downarrow\rangle$ states of System II are the same, the phase fluctuations caused by System II through H_1 are averaged and effectively becomes zero when $\gamma/J \gg 1$. When $\gamma/J < 1$, the oscillations caused by the interaction between System I and II are seen, as shown in figure 2.

In order to classify relaxations, we define the trace distance $D(\rho_a(t), \rho_b(t))$ between the state $\rho_a(t)$ and $\rho_b(t)$ of a qubit whose initial states are $\rho_a(0)$ and $\rho_b(0)$, respectively.

$$D(\rho_a(t), \rho_b(t)) \equiv \frac{1}{2} \text{Tr}(\sqrt{(\rho_a(t) - \rho_b(t))^\dagger (\rho_a(t) - \rho_b(t))}). \quad (11)$$

If there exists a pair of initial states $\rho_a(0)$ and $\rho_b(0)$ such that the time derivative of the trace distance between $\rho_a(t)$ and $\rho_b(t)$ at a certain t becomes positive, the noise process is called non-Markovian [1, 32, 33]. From these perspectives, we interpret the γ dependence of $|w(t)|$ shown in figure 2(b), as follows.

We define $\rho_I(t)$ as the density matrix at t when System II is traced out. In the above case, we find

$$\text{Tr}((\sigma_x + i\sigma_y)\rho_I(t)) = w(t).$$

The initial state at $t = 0$ gives $w(0) = 1$, while the state at $t = \infty$ gives $w(\infty) = 0$ since $\rho_I(\infty) = \sigma_0/2$. We assign $\rho_I(t)$ obtained the above to $\rho_a(t)$ in equation (11) and do $\sigma_0/2$ to $\rho_b(t)$. Note that the density matrix $\sigma_0/2$ corresponds to $(w_{\uparrow}, w_{\downarrow}) = (z_{\uparrow}, z_{\downarrow}) = (0, 0)$ and thus $\sigma_0/2$ does not evolve in time according to equation (8). We find

$$D(\rho_I(t), \sigma_0/2) = \frac{1}{2}|w(t)|,$$

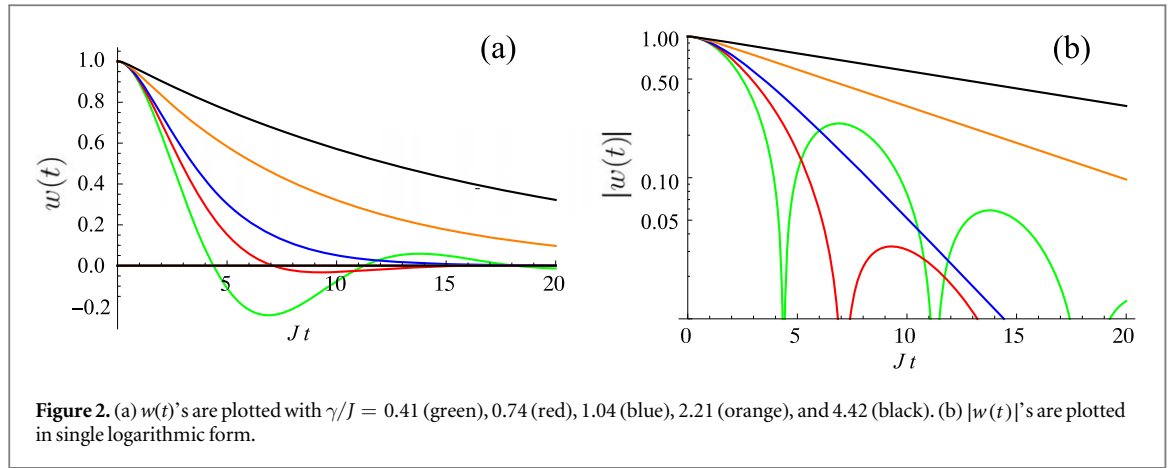


Figure 2. (a) $w(t)$'s are plotted with $\gamma/J = 0.41$ (green), 0.74 (red), 1.04 (blue), 2.21 (orange), and 4.42 (black). (b) $|w(t)|$'s are plotted in single logarithmic form.

or the FID signal directly shows the trace distance between $\rho_1(t)$ and $\sigma_0/2$. When γ/J is large, the relaxation is exponential, or $\frac{d \log D(\rho_1(t), \sigma_0/2)}{dt}$ is negative constant. This means that the time-homogeneous relaxation is observed. On the other hand, when γ/J is small, $\frac{dD(\rho_1(t), \sigma_0/2)}{dt}$ can be positive at a certain t . This implies that the relaxation is non-Markovian. When γ/J is intermediate, $\frac{d \log D(\rho_1(t), \sigma_0/2)}{dt}$ is always negative, but time-dependent. This implies that the time-inhomogeneous noisy environment is realized [1, 2, 34].

2.3. Non-Markovian environment with n qubits in System II

Let us consider the case when System II consists of n qubits. We call it a $(1 + n)$ system. We assume the following.

- There is no interaction among the qubits in System II.
- The individual interactions between the qubit in System I and the qubits in System II are the same as that in the $(1 + 1)$ system.
- The Markovian environments equally influence on the qubits in System II.
- The qubits in System II are distinguishable.

The master equation is given as,

$$\frac{d\rho_n}{dt} = \sum_{\ell=1}^n \left(-i \left[J \frac{\sigma_z \otimes \sigma_z^{(\ell)}}{4}, \rho_n \right] + \sum_{\pm} \gamma (2\mathcal{L}_{\mp}^{(\ell)} \rho_n \mathcal{L}_{\pm}^{(\ell)} - \{\mathcal{L}_{\pm}^{(\ell)} \mathcal{L}_{\mp}^{(\ell)}, \rho_n\}) \right), \quad (12)$$

where

$$\begin{aligned} \mathcal{L}_{\pm}^{(\ell)} &= \sigma_0 \otimes \sigma_{\pm}^{(\ell)}/2, \\ \sigma_k^{(\ell)} &= \sigma_0 \otimes \cdots \otimes \overset{\ell\text{th}}{\sigma_k} \otimes \cdots \otimes \sigma_0, \quad (k = x, y, z, +, -). \end{aligned}$$

On the other hand, the density matrix is $2^{n+1} \times 2^{n+1}$ matrix and is given as,

$$\rho_n = \sum_{k=\{\uparrow, \downarrow\}^n} \frac{x_k \sigma_x + y_k \sigma_y + z_k \sigma_z + \sigma_0}{2} \otimes \frac{|k\rangle \langle k|}{2^n}. \quad (13)$$

Note that $|k\rangle$ runs from $|\overbrace{\uparrow, \dots, \uparrow}^n\rangle, |\uparrow, \dots, \uparrow \downarrow\rangle, |\uparrow, \dots, \downarrow \uparrow\rangle, \dots, |\downarrow, \dots, \downarrow\rangle$. As in the $(1 + 1)$ system, z_k is constant because of $\sigma_z \otimes \sigma_z$ type interactions. Therefore, the dynamics of ρ_n is reduced to the simultaneous differential equations,

$$\frac{d\vec{w}_n}{dt} = M_n \vec{w}_n, \quad (14)$$

where $\vec{w}_n = (w_{\{\uparrow, \downarrow\}^n})^T$ has 2^n complex components and M_n is a $2^n \times 2^n$ matrix. By taking into account the structure of equation (12) and $M_1 = M$, M_n is given as,

$$\begin{aligned}
M_n &= M \otimes \sigma_0^{\otimes(n-1)} + \sigma_0 \otimes M \otimes \sigma_0^{\otimes(n-2)} + \dots + \sigma_0^{\otimes(n-1)} \otimes M \\
&= -\frac{n\gamma}{2} \sigma_0^{\otimes n} + \frac{\gamma}{2} \sum_{\ell} \sigma_x^{(\ell)} + \frac{i\gamma}{2} \sum_{\ell} \sigma_z^{(\ell)}.
\end{aligned} \tag{15}$$

M_n can be diagonalized with

$$V_n = V^{\otimes n}, \tag{16}$$

such that

$$\Lambda_n = \Lambda \otimes \sigma_0^{\otimes(n-1)} + \sigma_0 \otimes \Lambda \otimes \sigma_0^{\otimes(n-2)} + \dots + \sigma_0^{\otimes(n-1)} \otimes \Lambda. \tag{17}$$

2.4. Permutation symmetric non-Markovian environment

We relax the forth condition ‘The qubits in System II are distinguishable.’ In section 2.3 and consider the case when System II is symmetric with respect to permutation operations. In other words, what we have to consider is the dynamics in the symmetric subspace in the 2^n dimensional space spanned by the basis $|\{\uparrow, \downarrow\}^n\rangle$.

This permutation symmetric subspace is spanned by the following $1 + n$ vectors.

$$\left| \frac{n}{2}, \frac{n_{\uparrow} - n_{\downarrow}}{2} \right\rangle \equiv \sqrt{\frac{n_{\uparrow}! n_{\downarrow}!}{n!}} \sum_p P(|\overbrace{\uparrow \dots \uparrow}^{n_{\uparrow}} \overbrace{\downarrow \dots \downarrow}^{n_{\downarrow}}\rangle), \tag{18}$$

where \sum_p denotes summing all permutations. We make a $(1 + n) \times 2^n$ matrix

$$K_n = \left(\left| \frac{n}{2}, \frac{n}{2} \right\rangle, \left| \frac{n}{2}, \frac{n}{2} - 1 \right\rangle, \dots, \left| \frac{n}{2}, -\frac{n}{2} \right\rangle \right).$$

Then, M_n is projected on this subspace as follows.

$$M_n^{(s)} = K_n^T M_n K_n = -\frac{n\gamma}{2} I^{(n+1)} + \gamma S_x^{(n/2)} + i\gamma S_z^{(n/2)}, \tag{19}$$

where $S_k^{(m)}$ ($k = x, y, z$) is the spin- m matrix and $I^{(d)}$ is the identity matrix of dimension d . Transformation in equation (19) is based on the definition of $S_k^{(m)}$. Note that $M_n^{(s)}$ is a $(1 + n) \times (1 + n)$ matrix and is much smaller than M_n .

It is interesting to note that M_n and $M_n^{(s)}$ give the same dynamics of System I.

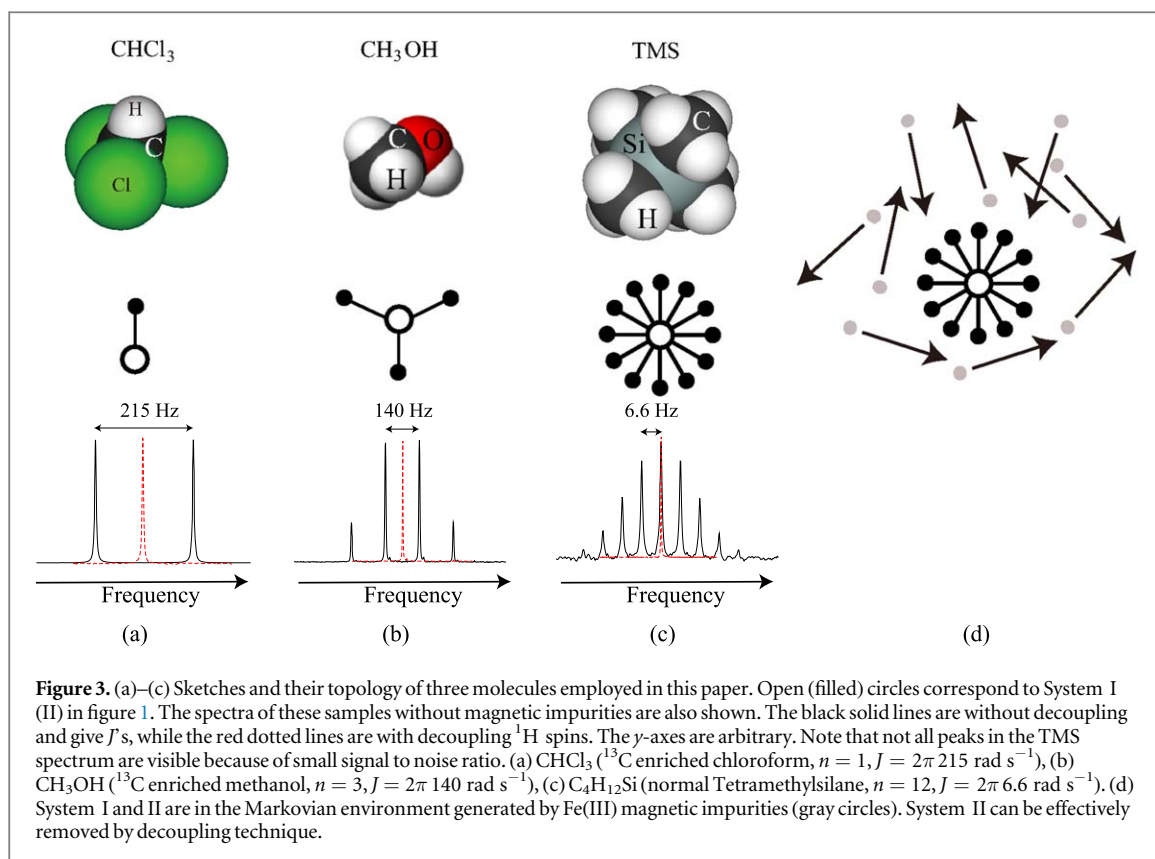
3. Experimental realization of non-Markovian environment

By using NMR, we demonstrate our idea to engineer the environment where System I interacts with a Markovian environment through System II in order to induce the time-inhomogeneous or non-Markovian dephasing. Solute molecules in isotropic liquids are under influence of solvent molecules. However, we have to take into account that these molecules are (1) under an external strong magnetic field, and (2) rapidly moving and rotating. Therefore, the interactions between nuclear spins in the solute molecules and solvent molecules of which time scales are much shorter than $1/\omega$ (ω is a Larmor frequency of the nuclear spin) are averaged out and are effectively nullified: this is the motional narrowing effect [36]. Moreover, if the solvent molecules do not have dipole moments, then the solute molecules’ nuclear spins can be approximately thought as in vacuum in the sense that the solvent, or the environment, does not have an influence on the nuclear spins in them [36]. Note also that the interactions among hetero-nuclear spins in a molecule solved in isotropic liquid are well described with $\sigma_z \otimes \sigma_z$ type ones [36] that are assumed in section 2.

Now, we add a small and controlled amount of ions that have magnetic dipole moments, such as Fe(III), into solution. We call these ions magnetic impurities. These magnetic impurities are moving rapidly and randomly because of the thermal motion, and thus they randomly flip-flop the solute molecules’ nuclear spins. So the magnetic impurities play the role of the environment that generates random flip-flop motions of the nuclear spins in solute molecules. The rate of these flip-flopping, γ , is proportional to the concentration of the magnetic impurities [21]. Note that the concentration of the magnetic impurities are controlled so that the magnetic impurities do not show a motional narrowing effect, see figure 2. Moreover, these magnetic impurities are rapidly running in the solution and thus this ‘environment’ has very short memory. Or, the magnetic impurities act as almost ideal Markovian environment. Although adding magnetic impurities for controlling longitudinal relaxations is a popular technique in NMR [36], the originality of our approach is that we use magnetic impurities to engineer the environment for realizing time-inhomogeneous and non-Markovian environment.

3.1. Samples

As the solute molecule for the present study, we employed (a) ^{13}C enriched chloroform (CHCl_3), (b) ^{13}C enriched methanol (CH_3OH), and (c) normal Tetramethylsilane (TMS, $\text{C}_4\text{H}_{12}\text{Si}$) in acetone d-6. Note that the



spins in Cl atoms in chloroform is magnetically inert, while that in O atom and those in C atoms in TMS are spinless. In these molecules, the ^{13}C or ^{29}Si spins (open circles in figure 3) correspond to System I, while the ^1H spins (filled circles) correspond to System II. About 5% of Si atoms in TMS is ^{29}Si and has a spin $1/2$. The qubit number n 's of system II are 1 (a), 3 (b), and 12 (c), while the interaction strengths J 's are $2\pi \ 215 \text{ rad s}^{-1}$ (a), $2\pi \ 140 \text{ rad s}^{-1}$ (b), and $2\pi \ 6.6 \text{ rad s}^{-1}$ (c) [21]. These J 's were measured from the separations of the peaks in the spectra (figure 3) obtained by Fourier transforming Free Induction Decay (FID) signals. System I and II are surrounded by the Markovian environment generated by Fe(III) magnetic impurities (gray circles), see figure 3(d). When the ^1H spins are decoupled, these spectra show only single peaks and indicate that the ^1H spins were effectively nullified. See section 3.2, too. Note that when the ^1H spins are decoupled, the Larmor frequencies of the ^{13}C and ^{29}Si spins in TMS are employed as the frequency standards [37]. Therefore, TMS is one of the most popular compounds in NMR.

As we mentioned above, these molecules are rotating in solutions and the spins consisting of System II are changing their positions. Therefore, the permutation symmetric non-Markovian environment model discussed in section 2.4 must be employed for analyzing experimental data. Moreover, the Markovian environment directly influences System I although it is smaller than that on System II because of the strong distance dependence of the dipole–dipole interaction: System I is farther from the magnetic impurities than System II, as schematically shown in figure 3(d). We may estimate the shortest distance between System I (II) and the magnetic impurity, as follows. The magnetic moment of the ion is caused by unpaired electron on the electrical orbit and thus it is reasonable to assume that the magnetic moment is on the surface of the ion. Therefore, the distance between System I and the magnetic moment is of the order of the sum of a Si atom radius (110 pm) and a ^1H atom one (50 pm) and is of the order of 150 pm. On the other hand, this distance between System II and the magnetic moment is of the order of the ^1H atom radius (50 pm).

From equation (4), the direct influence on System I from the magnetic impurities can be taken into account by introducing the following M'_n .

$$M'_n = -\frac{\gamma_0}{2} \sigma_0^{\otimes n} + M_n, \quad (20)$$

where γ_0 is the flip-flopping rate of System I directly caused by the Markovian environment. See appendix for more details.

3.2. Switching off non-Markovianity

The origin of the non-Markovian behavior of System I is System II. Therefore, if we were able to remove System II, it might be possible to switch off the non-Markovianity of the environment [31]. Of course, we cannot physically separate the ^1H atoms from the molecules shown in figure 3. However, the influence from the ^1H spins can be nullified by using a standard NMR technique called decoupling [36, 37].

We employed the WALTZ-16 pulse sequence [37] for decoupling. See, figure 4. The WALTZ-16 pulse sequence consists of multiple series of $+\pi/2$, $-\pi$, and $+3\pi/2$ -pulses without time intervals between them unlike usual decoupling pulses in quantum information processing [3–5]. We also note that usual NMR measurements are ensemble ones and that the system dynamics during the decoupling can be measured continuously. The strength of these pulses in our experiments were $H_{\text{rf}} = 2\pi \cdot 2.7 \times 10^3 \text{ rad s}^{-1}$ in frequency unit or the ^1H spins rotated at H_{rf} under these pulses. Since the interaction strengths of our samples are at most $2\pi \cdot 215 \text{ rad s}^{-1}$ in frequency unit, they are much less than H_{rf} . Therefore, the ^1H spins are effectively nullified in average. These nullification can be confirmed from the fact that the spectra contains only single peaks, as shown in figure 3, when the ^1H spins are decoupled with the WALTZ-16. Remember that we measured J 's from the peak separations. When the ^1H spins are nullified, γ_0 can be directly measured. We also note that there is no principle limitation in H_{rf} strength and one can increase it if necessary.

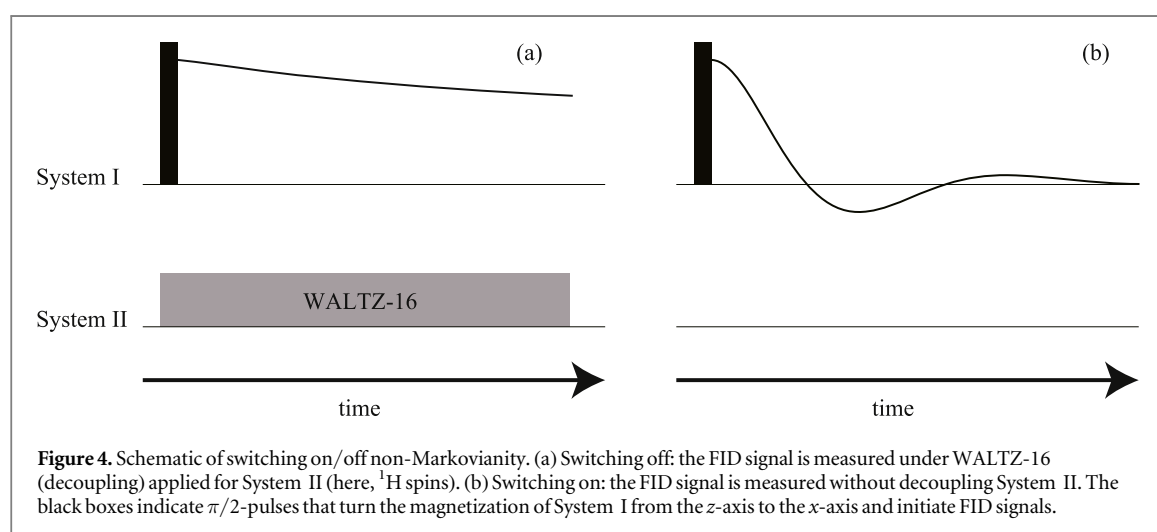


Figure 4. Schematic of switching on/off non-Markovianity. (a) Switching off: the FID signal is measured under WALTZ-16 (decoupling) applied for System II (here, ^1H spins). (b) Switching on: the FID signal is measured without decoupling System II. The black boxes indicate $\pi/2$ -pulses that turn the magnetization of System I from the z -axis to the x -axis and initiate FID signals.

3.3. (1 + 1) system

The left panels in figure 5 shows the measured Free Induction Decay signals (hereafter FID's) of ^{13}C in ^{13}C enriched CHCl_3 in acetone- d_6 when H spins were decoupled. The red (black) points are the real (imaginary) part of FID's. The dashed lines are exponential fittings to the real parts of FID's and we obtained $\gamma_0/2$'s as their time constants. We also independently measured T_1 's of H spins with the standard NMR technique called the inverse-recovery method [36, 37]. These T_1 's are assigned to γ 's in theoretical calculations with a (1 + 1) system. These are summarized in table 1.

The right panels in figure 5 shows FID's of ^{13}C when H spins were not decoupled, or with non-Markovianity. The green (blue) lines are theoretically calculated real (imaginary) parts of FID's with a (1 + 1) system. There is an excellent agreement between the measured and calculated FID's. Note that there is no fitting parameters except for the signal amplitude normalization.

Table 1. Chloroform parameters. C_m 's are the concentrations of Fe(III) impurity in milli Mol unit (mM). γ_0 's were measured from FID signals when ^1H spins were decoupled, as shown in the left panels in figure 5. T_1 's of ^1H spins were independently measured with the standard NMR technique (the inverse-recovery method). $J = 2\pi \cdot 215 \text{ rad s}^{-1}$.

C_m	$1/(\gamma_0/2)$	T_1 of H
4 mM	0.17 s	0.21 s
48 mM	0.10 s	6.5 ms

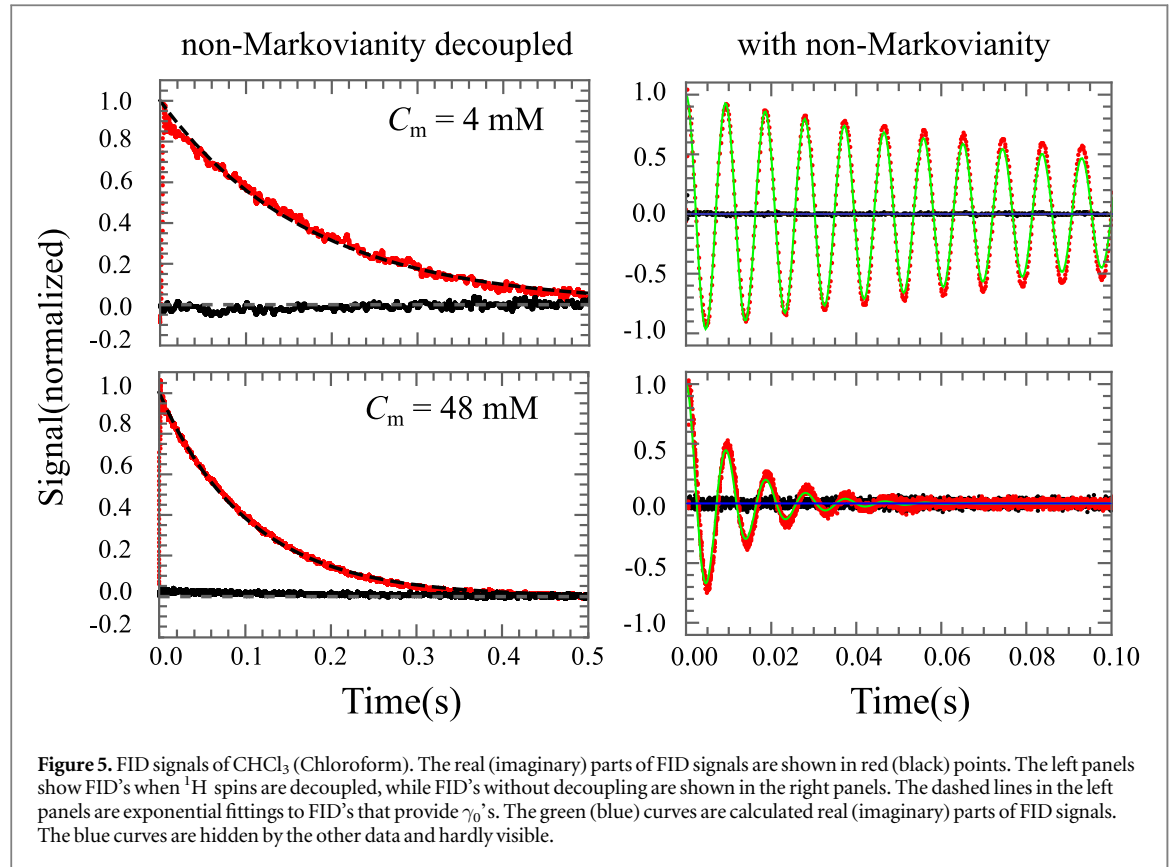


Table 2. Methanol parameters. The measurement methods are the same as in table 1.
 $J = 2\pi \cdot 140 \text{ rad s}^{-1}$.

C_m	$1/(\gamma_0/2)$	T_1 of H
12 mM	0.39 s	0.16 s
21 mM	0.37 s	93 ms
49 mM	0.10 s	34 ms
79 mM	0.13 s	20 ms

3.4. (1 + 3) system

Figure 6 shows the measured FID's of the CH_3OH samples and calculated ones with a (1 + 3) system like figure 5. The parameters for calculations are summarized in table 2. The slight increase of $1/(\gamma_0/2)$ from $C_m = 49$ to 79 mM might be caused by the motional narrowing effect. However, it is not straightforward to understand this quantitatively, which is kept as future research. Excellent agreement between the measured and calculated FID's are seen in figure 6, too. Note also that a close look (the inset of $C_m = 12$ mM) reveals that FID's without decoupling are not simple damping oscillations. This is an evidence that the environment has a long memory as we will discuss in section 3.5 in more detail. We also observe that the envelope in the non-decoupled case of $C_m = 79$ mM decays faster than that of $C_m = 49$ mM in spite of the longer $1/(\gamma_0/2)$ of $C_m = 79$ mM than that of $C_m = 49$ mM. The faster decay of the envelope is caused by the shorter T_2 of H, or larger γ in equation (12). This observation enforces our idea: the role of System II in the phase relaxation of System I is important.

3.5. (1 + 12) system

The excellent agreement between measured and calculated FID's shown in figures 5 and 6 illustrates the usefulness of our model to understand an open system with the environment that has a long memory. When n increases in a (1 + n) system, this long memory is more evident: we will discuss it with a (1 + 12) system in terms of an information (in our case, the transversal magnetization) flow [22].

Figure 7 shows FID's of ^{29}Si in TMS solved in acetone- d_6 and those calculated with a (1 + 12) system. The parameters are summarized in table 3. We again observed that γ_0 are not monotonous function of C_m . We, however, point out that the calculated FID signals with these measured γ_0 's reproduced our measured FID ones

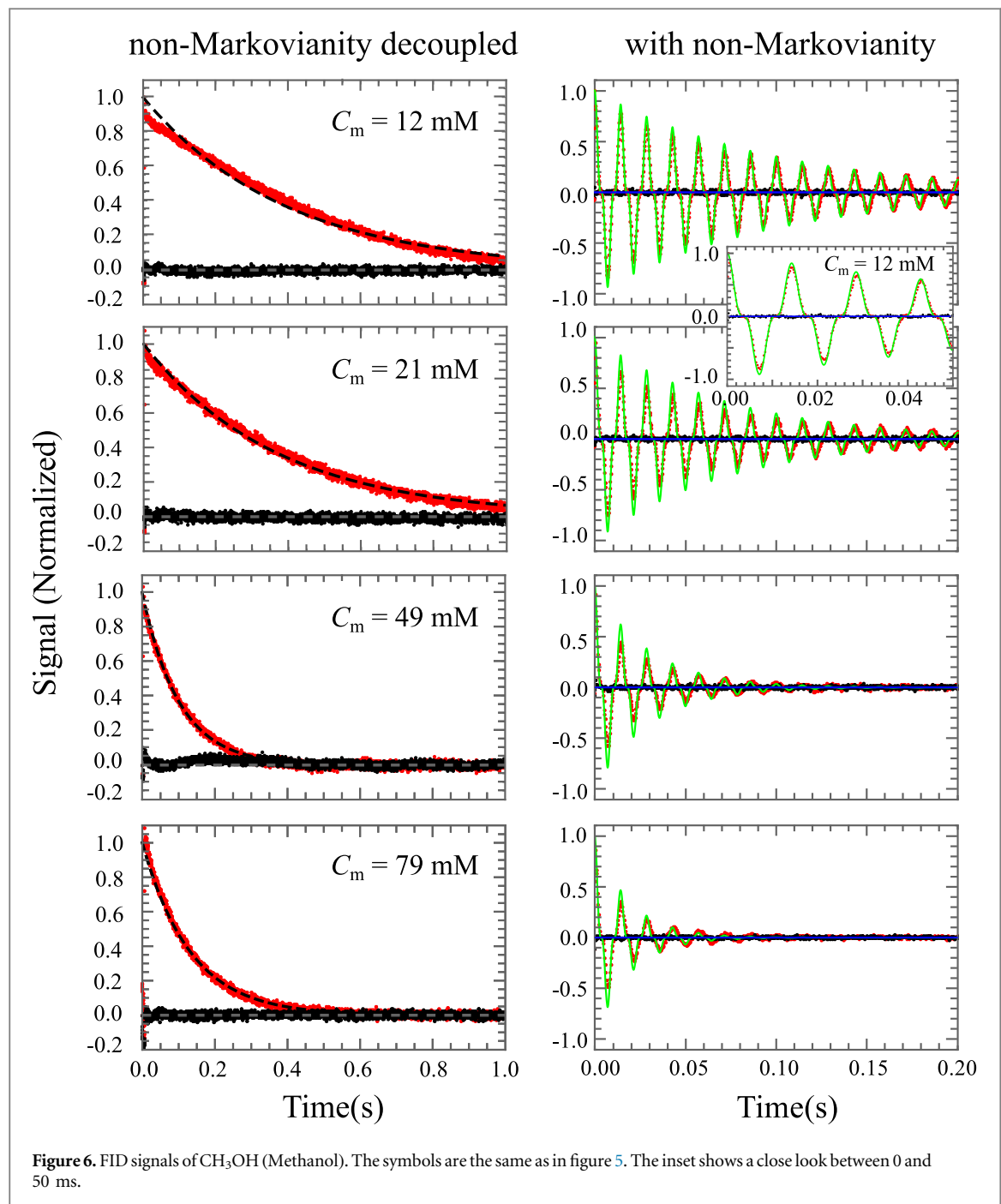


Table 3. TMS parameters. The measurement methods are the same as in table 1.
 $J = 2\pi \cdot 6.6 \text{ rad s}^{-1}$.

C_m	$1/(\gamma_0/2)$	T_1 of H
0 mM	4.9 s	10 s
11 mM	0.79 s	0.30 s
19 mM	1.0 s	0.14 s
40 mM	0.83 s	70 ms

well, as shown in figure 7. The measured FID signals in the cases of $C_m = 0$ and 11 mM appear noisier than those of $C_m = 19$ and 40 mM: this difference is caused by the differences of the number of measurements accumulated in order to improve the signal to noise ratio.

There might be small oscillations in the FID signals of $C_m = 0.0$ and 11 mM with non-Markovianity. We, however, think that these are within our experimental errors and thus these are not significant.

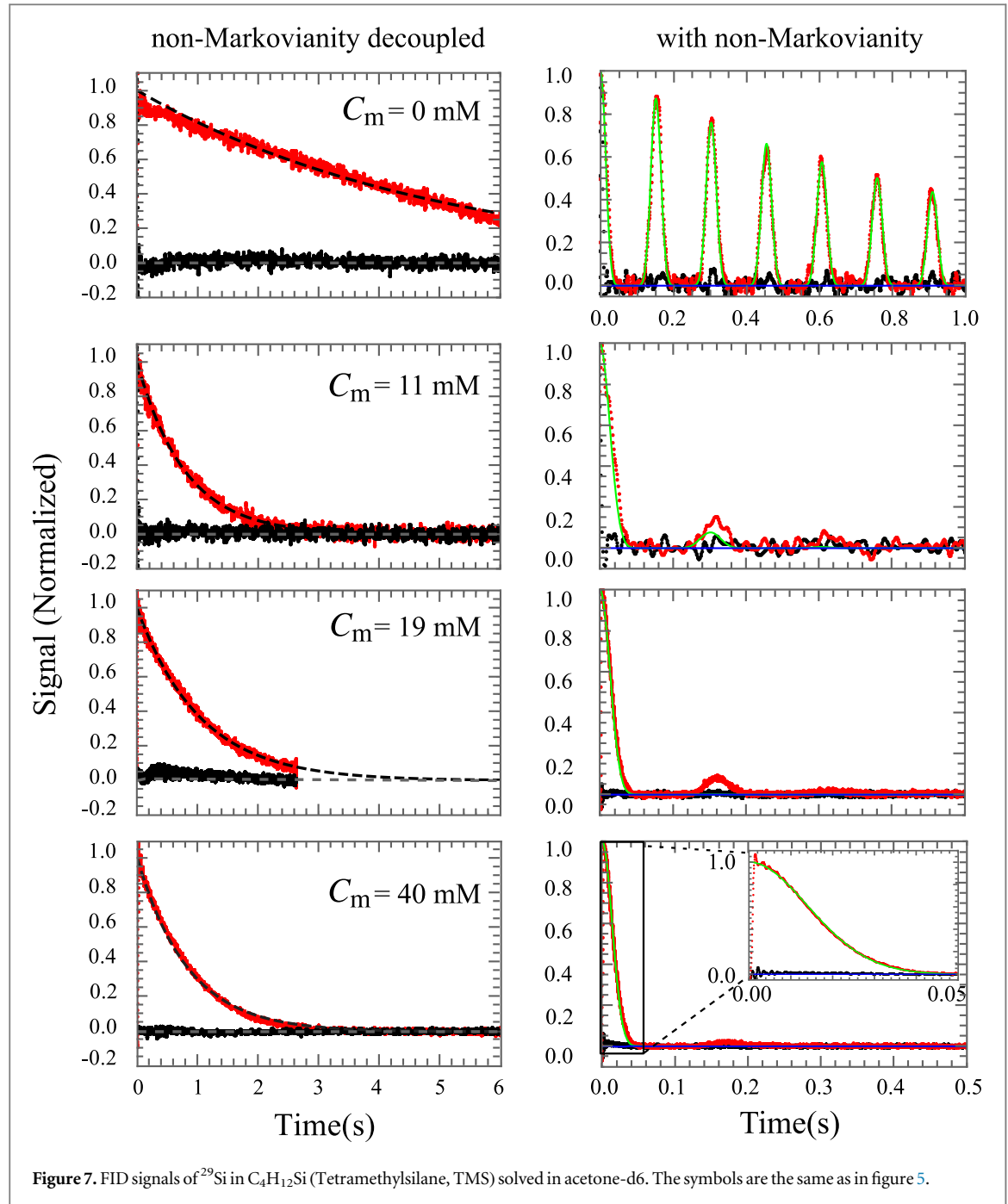


Figure 7. FID signals of ^{29}Si in $\text{C}_4\text{H}_{12}\text{Si}$ (Tetramethylsilane, TMS) solved in acetone- d_6 . The symbols are the same as in figure 5.

The FID signals (0 mM Fe(III)) with (left panel) and without (right panel) decoupling ^1H spins clearly show our ability of switching on and off non-Markovian environment. The time derivative of the trace distance $\frac{dD(\rho_1(t), \sigma_0/2)}{dt}$ becomes positive at a certain time t , and so this is the non-Markovian relaxation. In terms of the information flow, we can interpret this behavior as follows. The information flows from System I to System II and System I perfectly loses it at $t = 0.08$ s. The reverse flow, however, occurs and System I regains it at $t = 0.16$ s. This losing and regaining the information repeated many times. In other words, when System I loses the information, System II keeps it: this is the clear evidence that our engineered environment has a long memory.

On the other hand, the FID signals (40 mM Fe(III)) clearly show our ability of switching on and off time-inhomogeneous environment: The decay shown in the inset is non-exponential and $\frac{d \log(D(\rho_1(t), \sigma_0/2))}{dt}$ is always non-positive but its value is time-dependent. It originates from the time inhomogeneous noise. In terms of the information flow, we can interpret this behavior as follows. The information continuously flows from System I to (System II + the Markovian environment) and never reverses. Therefore, we may call it a Markovian process. Its flow rate, however, is time-dependent: this is the time-inhomogeneous relaxation.

The non-Markovian environment gradually changes to Markovian environment (but is time-inhomogeneous) in increasing the magnetic impurity concentration, as seen from the FID's of the sample with 11 and 19 mM Fe(III).

From the view point of the information flow, a $(1 + n)$ system must show a Markovian relaxation without the Markovian environment outside of System II if $n \rightarrow \infty$, or System II has infinite capacity of storing information [22].

4. Conclusion

In conclusion, we proposed a simple and practical model that shows time-(in)homogeneous Markovian and non-Markovian relaxations as a function of controllable parameters. Moreover, we experimentally demonstrate our idea with star topology molecules in isotropic liquids. The model consists of three parts: System I to be observed, System II surrounding System I and providing the memory of the interaction between System I and II, and Markovian environment. The dynamics of this system is analyzed with the Lindblad master equation that can be solved analytically. There is an excellent agreement between theoretical calculations and experimental results. Our model is useful to understand an open system, and this is essential for the realization of various quantum mechanics based technology, such as quantum computation and quantum meteorology.

Acknowledgments

This work is supported by CREST(JPMJCR1774) JST and in part by MEXT Grants-in-Aid for Scientific Research on Innovative Areas 'Science of hybrid quantum systems' (Grant No. 15H05870). YK is very grateful for Mikio Nakahara for useful discussions.

Appendix

We discuss a $(1 + 2)$ system that includes the direct influence on System I from Markovian environment in order to illustrate our idea of the permutation symmetric non-Markovian environment. The master equation is given as,

$$\begin{aligned} \frac{d\rho_2}{dt} = & \sum_{\pm} \gamma_0 (2 \mathcal{L}_{\mp}^{(0)} \rho_2 \mathcal{L}_{\pm}^{(0)} - \{\mathcal{L}_{\pm}^{(0)} \mathcal{L}_{\mp}^{(0)}, \rho_2\}) \\ & + \sum_{\ell=1}^2 \left(-i \left[J \frac{\sigma_z \otimes \sigma_z^{(\ell)}}{4}, \rho_2 \right] + \sum_{\pm} \gamma (2 \mathcal{L}_{\mp}^{(\ell)} \rho_2 \mathcal{L}_{\pm}^{(\ell)} - \{\mathcal{L}_{\pm}^{(\ell)} \mathcal{L}_{\mp}^{(\ell)}, \rho_2\}) \right), \end{aligned} \quad (21)$$

where γ_0 is the flip-flopping rate of System I directly caused by the Markovian environment and $\mathcal{L}_{\pm}^{(0)} = \sigma_{\pm} \otimes \sigma_0^{\otimes 2}/2$. On the other hand, the density matrix is $2^3 \times 2^3$ matrix and is given as,

$$\begin{aligned} \rho_2 = & \frac{1}{2} \sum_{k,l=\uparrow,\downarrow} \begin{pmatrix} 1 + z_{kl} & w_{kl}^* \\ w_{kl} & 1 - z_{kl} \end{pmatrix} \otimes \frac{|kl\rangle\langle kl|}{2^2} \\ = & \frac{1}{8} \begin{pmatrix} 1 + z_{\uparrow\uparrow} & 0 & 0 & 0 & w_{\uparrow\uparrow}^* & 0 & 0 & 0 \\ 0 & 1 + z_{\uparrow\downarrow} & 0 & 0 & 0 & w_{\uparrow\downarrow}^* & 0 & 0 \\ 0 & 0 & 1 + z_{\downarrow\uparrow} & 0 & 0 & 0 & w_{\downarrow\uparrow}^* & 0 \\ 0 & 0 & 0 & 1 + z_{\downarrow\downarrow} & 0 & 0 & 0 & w_{\downarrow\downarrow}^* \\ w_{\uparrow\uparrow} & 0 & 0 & 0 & 1 - z_{\downarrow\downarrow} & 0 & 0 & 0 \\ 0 & w_{\uparrow\downarrow} & 0 & 0 & 0 & 1 - z_{\uparrow\downarrow} & 0 & 0 \\ 0 & 0 & w_{\downarrow\uparrow} & 0 & 0 & 0 & 1 - z_{\downarrow\uparrow} & 0 \\ 0 & 0 & 0 & w_{\downarrow\downarrow} & 0 & 0 & 0 & 1 - z_{\uparrow\uparrow} \end{pmatrix} \\ = & \frac{1}{2^3} \begin{pmatrix} D_2^{(+)} & W_2^* \\ W_2 & D_2^{(-)} \end{pmatrix}. \end{aligned} \quad (22)$$

Next, substituting ρ_2 into the rhs of equation (21) gives

$$\frac{1}{16} \begin{pmatrix} A_{11} & 0 & 0 & 0 & A_{51}^* & 0 & 0 & 0 \\ 0 & A_{22} & 0 & 0 & 0 & A_{62}^* & 0 & 0 \\ 0 & 0 & A_{33} & 0 & 0 & 0 & A_{73}^* & 0 \\ 0 & 0 & 0 & A_{44} & 0 & 0 & 0 & A_{84}^* \\ A_{51} & 0 & 0 & 0 & -A_{11} & 0 & 0 & 0 \\ 0 & A_{62} & 0 & 0 & 0 & -A_{22} & 0 & 0 \\ 0 & 0 & A_{73} & 0 & 0 & 0 & -A_{33} & 0 \\ 0 & 0 & 0 & A_{84} & 0 & 0 & 0 & -A_{44} \end{pmatrix},$$

where

$$\frac{1}{16} \begin{pmatrix} A_{51} \\ A_{62} \\ A_{73} \\ A_{84} \end{pmatrix} = \frac{1}{8} \begin{pmatrix} iJ - \frac{1}{2}(\gamma_0 + 2\gamma) & \frac{\gamma}{2} & \frac{\gamma}{2} & 0 \\ \frac{\gamma}{2} & -\frac{1}{2}(\gamma_0 + 2\gamma) & 0 & \frac{\gamma}{2} \\ \frac{\gamma}{2} & 0 & -\frac{1}{2}(\gamma_0 + 2\gamma) & \frac{\gamma}{2} \\ 0 & \frac{\gamma}{2} & \frac{\gamma}{2} & -iJ - \frac{1}{2}(\gamma_0 + 2\gamma) \end{pmatrix} \begin{pmatrix} w_{\uparrow\uparrow} \\ w_{\uparrow\downarrow} \\ w_{\downarrow\uparrow} \\ w_{\downarrow\downarrow} \end{pmatrix}.$$

Note that the matrix in the rhs is equal to M'_2 introduced in equation (20). Therefore, we obtain

$$\frac{d\vec{w}_2}{dt} = M'_2 \vec{w}_2,$$

where $\vec{w}_2 = (w_{\uparrow\uparrow}, w_{\uparrow\downarrow}, w_{\downarrow\uparrow}, w_{\downarrow\downarrow})^T$.

Let us consider the case when the two qubits in System II are symmetric, i.e. $|\uparrow\downarrow\rangle$ and $|\downarrow\uparrow\rangle$ are indistinguishable. We introduce the Dicke basis as

$$|1, 1\rangle = |\uparrow\uparrow\rangle = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix}, \quad |1, 0\rangle = \frac{|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle}{\sqrt{2}} = \begin{pmatrix} 0 \\ \frac{1}{\sqrt{2}} \\ \frac{1}{\sqrt{2}} \\ 0 \end{pmatrix}, \quad |1, -1\rangle = |\downarrow\downarrow\rangle = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 1 \end{pmatrix},$$

and thus K_2 is given as

$$K_2 \equiv (|1, 1\rangle, |1, 0\rangle, |1, -1\rangle) = \begin{pmatrix} 1 & 0 & 0 \\ 0 & \frac{1}{\sqrt{2}} & 0 \\ 0 & \frac{1}{\sqrt{2}} & 0 \\ 0 & 0 & 1 \end{pmatrix}.$$

Symmetrized $W_2^{(s)}$ is obtained as follows.

$$W_2^{(s)} = K_2^T W_2 K_2 = \begin{pmatrix} w_{\uparrow\uparrow} & 0 & 0 \\ 0 & \frac{w_{\uparrow\downarrow} + w_{\downarrow\uparrow}}{2} & 0 \\ 0 & 0 & w_{\downarrow\downarrow} \end{pmatrix}.$$

Then, we can denote: $w_1 = w_{\uparrow\uparrow}$, $w_0 = \frac{w_{\uparrow\downarrow} + w_{\downarrow\uparrow}}{2}$, and $w_{-1} = w_{\downarrow\downarrow}$, respectively.

Now, we obtain the symmetrized $M_2^{(s)}$ by applying K_2^T and K_2 onto M'_2 , as follows.

$$\begin{aligned} M_2^{(s)} &= K^T M'_2 K = \begin{pmatrix} iJ - \frac{1}{2}(\gamma_0 + 2\gamma) & \frac{\gamma}{\sqrt{2}} & 0 \\ \frac{\gamma}{\sqrt{2}} & -\frac{1}{2}(\gamma_0 + 2\gamma) & \frac{\gamma}{\sqrt{2}} \\ 0 & \frac{\gamma}{\sqrt{2}} & -iJ - \frac{1}{2}(\gamma_0 + 2\gamma) \end{pmatrix} \\ &= -\frac{1}{2}(\gamma_0 + 2\gamma)I^{(3)} + iJS_z^{(1)} + \gamma S_x^{(1)}. \end{aligned}$$

Therefore, the dynamics of ρ_2 can also be obtained by solving

$$\frac{d}{dt} \begin{pmatrix} w_1 \\ w_0 \\ w_{-1} \end{pmatrix} = M_2^{(s)} \begin{pmatrix} w_1 \\ w_0 \\ w_{-1} \end{pmatrix}. \quad (23)$$

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