Persistent dynamic entanglement from classical motion: how bio-molecular machines can generate nontrivial quantum states

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Abstract. Very recently (Cai et al 2010 Phys. Rev. E 82 021921), a simple mechanism was presented by which a molecule subjected to forced oscillations, out of thermal equilibrium, can maintain quantum entanglement between two of its quantum degrees of freedom. Crucially, entanglement can be maintained even in the presence of very intense noise, so intense that no entanglement is possible when the forced oscillations cease. This mechanism may allow for the presence of nontrivial quantum entanglement in biological systems. Here we significantly enlarge the study of this model. In particular, we show that the persistent generation of dynamic entanglement is not restricted to the bosonic heat bath model, but can also be observed in other decoherence models, e.g. the spin gas model, and in non-Markovian scenarios. We also show how conformational changes can be used by an elementary machine to generate entanglement even in unfavorable conditions. In biological systems, similar mechanisms could be exploited by more complex molecular machines or motors.

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1. Introduction

The last couple of years have witnessed the birth and explosive development of a new research area, namely quantum biology. For many years, the possibility of nontrivial quantum effects in biological systems was thought to be impossible due to the large amount of noise that characterizes such systems. During the last few years however, very surprisingly, a host of experimental and theoretical works have pointed to the possible presence of such effects. Notable results concern the existence of quantum coherence in photosynthesis [1–17], quantum effects in the so-called ‘chemical compass’ [18–23] and phonon-assisted electron tunneling in the olfactory mechanism [24–26].

Of particular interest is the possible existence of nontrivial entanglement in biological systems. The reason is that entanglement has dramatic effects; in particular, it leads to a vastly increased capacity for information processing. Indeed, entanglement is at the core of quantum computation. The capacity of quantum systems to perform many other tasks is also significantly increased by entanglement. Given these benefits it is very tempting to suspect that biological systems may make use of entanglement. On the other hand, the existence of entanglement in biological systems was thought to be even more unlikely than that of mere quantum coherence because entanglement is extremely fragile and almost impossible to survive in the presence of noise. Very recently, however, it was realized [27] that the intuition about the fragility of entanglement was based on considering the behavior of quantum systems at thermal equilibrium. Biological systems, however, are not at equilibrium—they are open-driven systems, far from thermal equilibrium. Such systems are capable of ‘error correction’ and may actively maintain entanglement in the presence of noise. This raises the possibility that nontrivial entanglement may actually be present in biological systems.

In [28], a simple mechanism was presented by which a molecule subjected to forced oscillations, out of thermal equilibrium, can maintain quantum entanglement between two of its quantum degrees of freedom. Crucially, entanglement can be maintained even in the presence of very intense noise, so intense that no entanglement is possible when the forced
Figure 1. Conformational changes of a bio-molecule [29], induced e.g. by the interaction with some other chemical, can lead to a time-dependent interaction between different sites (blue circles) of the molecule. Also see [27].

oscillations cease. A potential biological system in which such an effect may take place is that of complex molecules (such as proteins) undergoing forced conformational changes. To date, no such system has been experimentally observed, for the simple reason that no experiment has so far been carried out to look for it. To be able to do that, it is imperative to first gain a better theoretical understanding of the model. This is the focus of this paper.

The paper is organized as follows. In section 2, we present the abstract model of a moving ‘two-spin molecule’ with a time-dependent coherent Hamiltonian; then we investigate the system behavior and the entanglement dynamics under the influence of two distinct environments. In section 3, we consider a bosonic heat bath representative of a large number of harmonic oscillators, and study its influence in both the Markovian and non-Markovian regimes. To relax the working assumption of periodic oscillations, we further introduce stochasticity in the classical motion of the molecule, and include an additional source of noise to illustrate the robustness of the entanglement generation. In section 4, we extend our study to the spin gas model. Section 5 is devoted to some concluding remarks.

2. How conformational molecular motion can generate entanglement

In the following, we consider bio-molecular systems that undergo controlled conformational changes, using a semi-quantal picture. We assume that the conformational motion of the molecular structure can be described, to a very good approximation, classically. In addition, however, we identify selected quantum degrees of freedom, embedded in this ‘backbone structure’ and localized at several classically defined sites, which interact with a strength depending on their distance (see figure 1).

In the following, we consider two quantum degrees of freedom which are forced to move along some predetermined trajectories and interact via dipolar coupling. Each of these degrees of freedom shall be represented by a quantum mechanical two-level system. For easy reference, we call such a model system a ‘two-spin molecule’ [28]. It can be regarded as an elementary example of a molecular machine which, as we shall see, is able to persistently generate entanglement. As such, it also presents a building block that could be exploited in more complex bio-molecular motors.

In more detail, the mechanism is described as follows: the conformational changes in the molecule force the two localized spins (or, more generally, two-level systems) to come close or move apart, inducing an effective time-dependent coupling, both between the two spins and between each spin and a local field, as described in figure 2. Furthermore, it will be assumed
Figure 2. A two-spin bio-molecule undergoes conformational changes such as harmonic oscillation or stochastic motion. Both the spin–spin interaction strength $J$ and the background field $B$ are position dependent. (Figure taken from [28].)

(this is essential) that the de-coherent environment is so noisy that no static entanglement between the two spins can exist in the static steady state, which is achieved when the molecular configuration is fixed to any of the possible ones. As we shall see in the following, if the molecular configuration oscillates in a periodic way, cyclic generation of fresh entanglement can persist even in the long-time limit. This feature will persist even in the presence of an additional source of dephasing, which would generally eliminate all coherence from the system. Finally, we shall generalize these considerations to more realistic scenarios involving non-Markovian environments and stochasticity in the classical motion.

In the following, whenever we refer to spins, we may imagine spin-1/2 magnetic moments interacting via a dipolar coupling and subjected to a background magnetic field. To conduct our study, we further assume that the two spins are coupled with the usual Ising-type interaction which depends only on the distance $d$ between the spins, and that there exists a local magnetic field which induces a (Zeeman) energy level splitting. The energy splitting is also assumed to depend only on the inter-spin distance $d$, which thus plays the role of an effective parameter for the overall molecular configuration. Explicitly, the Hamiltonian for the system is

$$H_M(d) = J(d)\sigma_1^{(1)}\sigma_2^{(2)} + B(d)(\sigma_1^{(1)} + \sigma_2^{(2)}),$$

where $\sigma_\alpha^{(\alpha)}$, $\sigma_\alpha^{(\alpha)}$ are Pauli operators of the $\alpha$th spin, $\omega_0(d) = 2B(d)$ is the local level splitting, and we set the Planck constant $\hbar = 1$. Since the molecule undergoes conformational changes, the distance $d$ between the two spins becomes a function of time: $d \equiv d(t)$. The explicit time dependence of $H_M(d) \equiv H_M(d(t))$ is determined by the specific choice of the molecular motion: initially, we consider forced periodic oscillations; later we include an additional stochasticity. With this model, the molecular motion leads to an effective time-dependent Hamiltonian for the system that we can write as

$$H_M(d(t)) \equiv H_M(t) = J(t)\sigma_1^{(1)}\sigma_2^{(2)} + B(t)(\sigma_1^{(1)} + \sigma_2^{(2)}).$$

Here and in the following, we use the short notation $J(t) \equiv J(d(t))$ for all the quantities that depend on distance $d$ and so, implicitly, on time $t$. For notational simplicity, we introduce the energy scale

$$E(t) = \sqrt{4B^2(t) + J^2(t)}$$

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in terms of which the eigenenergies of the system Hamiltonian $H_M(t)$ and the corresponding eigenstates can be written as

$$
\begin{align*}
\epsilon_0(t) &= -E(t), \quad |\epsilon_0(t)\rangle = (-\eta |00\rangle + |11\rangle)/(1 + \eta^2)^{1/2}, \\
\epsilon_1(t) &= -J(t), \quad |\epsilon_1(t)\rangle = (|01\rangle - |10\rangle)/\sqrt{2}, \\
\epsilon_2(t) &= +J(t), \quad |\epsilon_2(t)\rangle = (|01\rangle + |10\rangle)/\sqrt{2}, \\
\epsilon_3(t) &= +E(t), \quad |\epsilon_3(t)\rangle = (|00\rangle + \eta |11\rangle)/(1 + \eta^2)^{1/2}
\end{align*}
$$

with

$$
\eta \equiv \eta(t) = \frac{E(t) - 2B(t)}{J(t)}
$$

and $|0\rangle \equiv |\uparrow\rangle$, $|1\rangle \equiv |\downarrow\rangle$ being the eigenstates of $\sigma_z$ corresponding to the eigenvalues $+1$, $-1$, respectively. It can be seen that the energy spectra of the system Hamiltonian $H_M(t)$, and in particular the ground state $|\epsilon_0\rangle$, depend on both the interaction strength $J(t)$ and the local field $B(t)$ and thus are also time dependent. In the case of weak spin–spin interaction $J(t) \ll B(t)$, e.g. when two spins are spatially separated from each other, the system Hamiltonian can be approximated as $H_M \sim B(t)(\sigma_z^{(1)} + \sigma_z^{(2)})$, and the ground state approaches the separable state $|11\rangle$. As the spins move closer and the spin–spin interaction increases, the ground state changes according to $|\epsilon_0(t)\rangle$ in equation (4). In particular, the entanglement contained in the ground state $|\epsilon_0(t)\rangle$ is a monotonically increasing function of the ratio $J(t)/B(t)$, and can be quantified by the measure of concurrence $[30]$. Concurrence is zero for separable states and reaches its maximum value $C = 1$ for maximally entangled states; here it can be calculated as

$$
C(|\epsilon_0(t)\rangle) = \left| \langle \epsilon_0(t) | \sigma_y \otimes \sigma_y | \epsilon_0^* (t) \rangle \right| = 2\eta/(1 + \eta^2) = \left[ 1 + 4 \left( \frac{B(t)}{J(t)} \right)^2 \right]^{-1},
$$

where $|\epsilon_0^* (t)\rangle$ is the complex conjugate of state $|\epsilon_0(t)\rangle$ (expressed in the computational product basis, already used in equation (4)). In the meantime, the relative energy gap between the ground state and the first excited state $(\epsilon_1 - \epsilon_0)/(\epsilon_3 - \epsilon_0)$ decreases with the ratio $J(t)/B(t)$.

Before taking the environment into account, it is worth looking at the adiabatic coherent evolution of the oscillating molecule, which will facilitate our discussions in the following sections. If the system Hamiltonian changes slowly in time, the instantaneous eigenspaces of $H_M(t)$ will undergo independent evolution. The widely used condition that ensures the adiabatic dynamics for closed quantum systems is

$$
\left| \frac{\langle \epsilon_k(t) | \epsilon_l(t) \rangle}{\epsilon_k(t) - \epsilon_l(t)} \right| \ll 1, \quad k \neq l.
$$

For a detailed discussion concerning the validity of the adiabatic approximation, see $[31, 32]$. For the time-dependent system Hamiltonian introduced in equation (2), one easily gets

$$
|\epsilon_1(t)\rangle = |\epsilon_2(t)\rangle = 0
$$

and

$$
|\epsilon_0(t)\rangle = -\frac{\hat{\eta}}{1 + \eta^2} |\epsilon_3(t)\rangle, \quad |\epsilon_3(t)\rangle = \frac{\hat{\eta}}{1 + \eta^2} |\epsilon_0(t)\rangle.
$$

Thus the adiabatic condition can be explicitly expressed as

$$
\left| \frac{\hat{\eta}}{1 + \eta^2} \right| \ll 2E(t).
$$
Whenever the adiabatic condition is satisfied, the molecule that is initially in an eigenstate of the system Hamiltonian will remain in the corresponding eigenstate, and will acquire a dynamical, as well as geometric, phase:

\[ |\epsilon_k(0)\rangle \rightarrow U(t, 0) |\epsilon_k(0)\rangle \simeq e^{-i \int_0^t \epsilon_k(s) ds} e^{i \theta_k(t)} |\epsilon_k(t)\rangle, \]

where \( \int_0^t \epsilon_k(s) ds \) is the dynamical phase and \( \theta_k(t) = i \int_0^t \langle \epsilon_k(s) | \epsilon_k(s) \rangle ds \) represents the geometric phase [31]. One can check from equations (7) and (8) that \( \langle \epsilon_k(t) | \epsilon_k(t) \rangle = 0 \) is fulfilled \( \forall k \) and all times. Thus no geometric phase is accumulated, i.e. \( \theta_k(t) = 0 \) for all \( k \), and the adiabatic evolution can be expressed as follows:

\[ |\epsilon_k(0)\rangle \rightarrow e^{-i \int_0^t \epsilon_k(s) ds} |\epsilon_k(t)\rangle. \]

3. The bosonic thermal bath

The coherent part of the spin dynamics due to the time-dependent system Hamiltonian has to be incorporated into a more complex description which takes into account the presence of an environment. The actual environment of biological systems is very complex and there is in fact a large variety of environments (protein vibrations, solvent effects, spatially correlated environments, etc), which are different for the different systems under consideration: for a detailed quantitative analysis, one would have to take into account the specific nature of the environment. However, to gain first insights into the dynamics presented here, we adopt certain representative and well-investigated decoherence models. We first demonstrate the essential idea by considering the familiar decoherence model of spins coupled with independent thermal baths consisting of a large number of harmonic oscillators. The Hamiltonian of the individual thermal bath coupled to the \( \alpha \)th spin (\( \alpha = 1, 2 \)) is

\[ H_{SB}^{(\alpha)} = \sum_k \omega_k a_{k,\alpha}^\dagger a_{k,\alpha}, \]

where \( a_{k,\alpha}^\dagger \) and \( a_{k,\alpha} \) are the creation and annihilation operators for the \( k \)th bath oscillator mode and \( \omega_k \) the corresponding mode frequency. The coupling between the spin and the thermal bath is described by the following interaction Hamiltonian:

\[ H_{SB} = \sum_\alpha H_{SB}^{(\alpha)} = \sum_\alpha \sigma_x^{(\alpha)} \sum_k v_{k,\alpha} (a_{k,\alpha} + a_{k,\alpha}^\dagger), \]

where \( v_{k,\alpha} \) is the coupling amplitude to mode \( k \).

Within the Born–Markov approximation, and by assuming that the adiabatic condition for closed quantum systems equation (9) is satisfied, we have derived a time-dependent Lindblad-type quantum master equation [28] for the system dynamics under the rotating wave approximation, which reads as

\[ \frac{d}{dt} \rho(t) = -i[H_M(t), \rho(t)] + D(t)\rho(t) = L(t)\rho(t), \]

with the following dissipator:

\[ D(t)\rho(t) = \sum_{\alpha, \omega(t)} \Gamma_{\omega(t)} \{ A_\alpha[\omega(t)]\rho(t)A_\alpha^\dagger[\omega(t)] - A_\alpha^\dagger[\omega(t)]A_\alpha[\omega(t)]\rho(t) \} + \text{h.c.} \]

\[ A_\alpha[\omega(t)] = \sum_{\Delta \xi(t) = \omega(t)} \langle \epsilon_i(t) | \sigma_x^{(\alpha)} | \epsilon_j(t) \rangle \times | \epsilon_i(t) \rangle \langle \epsilon_j(t) |. \]
In other words, the time dependence of the system Hamiltonian introduces an implicit time dependence into the Lindblad generators \( A_j(t) \), as well as into the rates \( \Gamma_{\omega(t)} \) (a sort of one-sided Fourier transform of the thermal bath correlation function, see [28]). Note that, as already discussed for equation (2), the time dependence of \( H_d \) (and so also \( D \) and \( L \)) is actually due to its dependence on the distance \( d \) between the spins, from which arises the short notation, e.g. \( L(t) \equiv L(d(t)) \).

The bosonic thermal bath is characterized by its spectral density function \( J(\omega) \), which contains information on both the density of modes in the frequency domain and the strength of their coupling to the system. In the following, we choose an Ohmic spectral density \( J(\omega) = \frac{k}{\omega_c} \omega e^{-\omega/\omega_c} \) with cut-off frequency \( \omega_c \). The memory time of the thermal bath strongly depends on the cut-off frequency and so does its Markovian or non-Markovian behavior [33]. In particular, if \( \omega_c \gg \omega \), where \( \omega \) is the energy scale of the system transition frequencies, the reservoir loses its memory quickly, and is called Markovian. The corresponding correlation function exhibits a sharply peaked kernel in the limit \( \omega_c \rightarrow \infty \). In this case, one can simplify the spectral density as \( J(\omega) = \kappa \omega \) with \( \kappa = \frac{k}{2\pi} \). We can thus write the Fourier transformation of the thermal bath correlation function as \( \Gamma_{\omega(t)} = \kappa \omega(t)(1 + N_{\omega(t),\beta}) \), where \( N_{\omega(t),\beta} = 1/(e^{\omega(t)/\beta} - 1) \) represents the bosonic distribution function at inverse temperature \( \beta \).

In the following, we set the thermal energy \( k_B T \) as the unit of the energy scale, in this way also fixing \( \beta = 1 \). At room temperature, \( T = 300 \text{K} \) and the thermal energy \( k_B T \) is approximately 26 meV. In this case, an energy \( E = 2 \) actually means \( E = 2k_B T \approx 52 \text{meV} \).

In order to establish a comparison between the static and dynamic cases, we first consider the case when the two-spin bio-molecule is fixed in a static configuration. Since the two spins are coupled to independent thermal baths at the same temperature, the corresponding quantum master equation mixes the energy eigenstates in the sense that the molecule is driven toward its thermal equilibrium state [33] at the reservoir temperature \( 1/\beta \). For each spin–spin interaction strength \( J(d) = J \) and local magnetic fields \( B(d) = B \), the entanglement of the corresponding thermal equilibrium state, defined by \( \mathcal{L}(d) \rho_{th} = 0 \), is quantified by [28]

\[
C(\rho_{th}) = \frac{2}{Z} \max \left\{ 0, \frac{2\eta}{1 + \eta^2} \sinh (E\beta) - \cosh (J\beta) \right\},
\]

with \( Z = 2 \cosh (E\beta) + 2 \cosh (J\beta) \) being the partition function. After some calculations, it can be seen that the first derivative of \( C \) with respect to \( \beta \) is always non-negative

\[
\frac{\partial C(\rho_{th})}{\partial \beta} \geq 0,
\]

which means that the static thermal entanglement decreases as the temperature increases; in particular, static entanglement cannot survive if the temperature is higher than a certain critical temperature \( 1/\beta_c \). In other words, if the molecule stays in a fixed configuration, entanglement will finally disappear for high enough temperature; see, e.g., figure 3. In this sense, the bosonic heat bath, even though it may not be a very realistic model of a biological environment, plays a useful role in our investigation. It is an example of an unfavorable environment, which cannot introduce any entanglement by itself.

Here we address the key question: What happens if one takes into account the molecular motion? and in particular: Can entanglement possibly build up through the classical motions of the molecule? The answer is in the affirmative and we demonstrate in the following that entanglement can indeed persistently recur in an oscillating molecule, even if the environment is so hot that any static thermal state is separable, i.e. the temperature \( T > \max \{T_c\} \) for all possible molecular configurations encountered during the motion.
Figure 3. Static entanglement in the thermal equilibrium state at different inverse temperatures $\beta$ for $B(x) = B(0) - x$ and $J(x) = J(0) + x$ with $B(0) = 1.2$ and $J(0) = 0.1$. Here $\beta$, $B$, $J$ can be varied independently, but it is their relative value that determines the dynamical behavior. Therefore, here and in the following, we use dimensionless quantities obtained by setting the thermal energy $k_B T$ as the unit of the energy scale (so that $\beta = 1$) with respect to which all the other parameters are determined, for example $B = 1$ indicates that $\bar{h} B / k_B T = 1$.

3.1. Qualitative physical picture

Before proceeding, it is useful to present a simple qualitative picture which illustrates the idea of how entanglement can be established through classical motion even in noisy environments. We consider the following simple process: at time $t = 0$, two spins are far from each other and the spin–spin interaction is very weak compared to the local magnetic fields. Thus the system Hamiltonian can be approximated as $H_M(0) \simeq B(0)(\sigma_z^{(1)} + \sigma_z^{(2)})$. Due to the presence of the thermal bath, the molecule is driven into its thermal equilibrium state with the corresponding ground state population denoted as $p_0$. If the local energy level splitting $\omega_0(0) = 2B(0)$ is comparable to or larger than the thermal energy scale, $\hbar \omega_0(0) \gg k_B T$, the ground state population $p_0$ at this distant configuration will be relatively larger than the corresponding populations of the other energy levels. Nevertheless, entanglement does not appear since the ground state is separable ($|\epsilon_0(0)\rangle \simeq |11\rangle$). Let us further assume that the spins come closer at such a speed that the coherent evolution due to the system Hamiltonian is adiabatic. While the spins approach each other, their interaction becomes stronger and the energy eigenstates adiabatically change into the ones of $H_M(t)$. In particular, the ground state $|\epsilon_0(0)\rangle \rightarrow |\epsilon_0(t)\rangle$ becomes more and more entangled as the coupling strength between spins increases, as indicated by the fact that its concurrence $C(|\epsilon_0(t)\rangle) = J/\mathcal{E}$ is an increasing function of the ratio $J(t)/B(t)$. Following this reasoning, it is possible that entanglement appears as a transient phenomenon as long as the adiabatic change of $H_M$ is still fast compared to the thermalization time scale $\gamma^{-1}$ with $\gamma = \kappa \omega(2N_{\omega,\beta} + 1)$ for the chosen simple Ohmic spectral density.

To make the argument more concrete, in figure 4 we illustrate the above qualitative picture through a simple process, in which

$$B(t) = \begin{cases} B(0) - \delta \cdot t, & 0 \leq t \leq t_0, \\ B(t_0), & t_0 \leq t. \end{cases}$$

(18)
Figure 4. Entanglement $C$ (a) and the ground state population $p_0$ (b) versus $t$ for a single conformational change with $B(t)$ and $J(t)$ as in equations (18) and (19) and $B(0) = 1.2$, $J(0) = 0.1$, $\delta = 0.01$, $t_0 = 100$, with different system–bath coupling strengths. Here and in the following, the reservoir inverse temperature is $\beta = 1$.

\[
J(t) = \begin{cases} 
J(0) + \delta \cdot t, & 0 \leq t \leq t_0, \\
J(t_0), & t_0 < t.
\end{cases}
\tag{19}
\]

Considering a reservoir with inverse temperature $\beta = 1$, the initial ground state population $p_0$ is about 85%. The change of the system Hamiltonian is characterized by the small step value $\delta = 0.01$, which ensures the adiabatic condition for closed systems. It can be seen that the molecule can be kicked out of its separable thermal equilibrium state through classical motion and thus entanglement grows before being washed out by thermalization. The dissipative environment now plays its usual destructive role. Moreover, if the thermalization rate is fast compared to the adiabatic change of the system Hamiltonian, entanglement is greatly suppressed.

3.2. Persistent recurrence of fresh entanglement

To counteract the detrimental effect of the environment and to sustain the generation of entanglement as a nontransient phenomenon, a persistent supply of free energy is needed. This is an organized form of energy which, as in our mechanism, can be introduced by conformational changes. To illustrate our idea, we first consider a two-spin molecule subjected to a deterministic and periodic oscillatory motion (the strict periodicity will be relaxed in section 3.4). The position of the two spins is given by

\[
x_\alpha(t) = x_\alpha(0) + (-1)^\alpha a \left( \cos \frac{2\pi t}{\tau} - 1 \right), \quad \alpha = 1, 2,
\tag{20}
\]

where $x_\alpha(0)$ are the initial positions, $a$ is the amplitude of oscillation and $\tau$ is the oscillation period. The distance between the two spins is thus determined by

\[
d(t) = |x_1(t) - x_2(t)|.
\tag{21}
\]

As a remark, we refer to section 3.4 for more general forms of classical motion involving stochasticity. The coupling between the two spins is described by a dipole–dipole Ising
Figure 5. (a) Entanglement $C$ versus the molecular configuration $d$ (the distance between the two spins as defined in equation (21)) for the bosonic thermal bath with $\beta = 1$ and $\kappa = 0.01$. The dot-dashed, blue curve indicates the first period; the solid, red curve marks the asymptotic cycle; the green, dotted curve refers to the thermal equilibrium case. The arrows along the curves indicate how the entanglement evolves during the classical oscillations. The oscillation parameters are $x_1(0) = -x_2(0) = -20$, $a = 5$, $\tau = 100$ and $B_0 = 1.3$, $B_1 = 2.4$, $\sigma = 120$, $J_0 = 10^4$. (b) Entanglement during the asymptotic cycle for different system–bath coupling strengths.

The interaction

$$J(t) = J_0/d^3(t),$$

while the local magnetic field profile, dependent on the spin position, is chosen to be

$$B_\alpha(t) = B_0 - B_1 \exp^{-x_\alpha^2(t)/\sigma} = B_0 - B_1 \exp^{-d^2(t)/4\sigma}.$$ 

The choice of $x_\alpha(t)$, $B_\alpha(t)$ and $J(t)$ is quite arbitrary. As is explained in the following analysis, the essential condition for the proper function of the mechanism is that the dominant contribution to $H_M(t)$ is due to the background field when the spins are far away and due to their interaction when they are closest. This is a natural assumption, since the interaction generally increases when the distance is reduced.

We assume that the two spins start from the distant configuration and are initially in the corresponding thermal equilibrium state. Note that this particular initial condition is irrelevant for the system asymptotic dynamics that we are interested in. At time $t = 0$ the spins start to oscillate, i.e. they periodically come close to each other and separate again. The state of the molecule is driven out of thermal equilibrium, and entanglement is generated. When the spins move back to their distant configuration, the thermalization tends to increase the population of the ground state (due to the increasing energy gap between the ground state and the first excited state), and thus effectively resets the system to a favorable condition. Therefore, although environmental noise generally destroys entanglement, it now represents a crucial factor to enable the persistent and cyclic generation of fresh entanglement. For simplicity, we call the cycle during the time interval $[n\tau, (n+1)\tau]$ with $n \rightarrow \infty$ the asymptotic cycle. It can be seen from figure 5 that entanglement indeed persistently recurs during the oscillations even if the thermal bath is so hot that no static entanglement can survive.

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In figure 6, we plot the population of the instantaneous ground state of the system Hamiltonian $H_M(t)$ during the oscillation and observe how the periodic oscillations keep the molecule far away from its thermal equilibrium (as compared with the green curve). In particular, when the spins get closer and the entanglement of the ground state increases, the ground state occupation is noticeably higher than the thermal one, and this actually constitutes the key to the mechanism. Let us point out once more that thermalization tends to drive the molecule close to its instantaneous equilibrium state, but the continuous change of its shape prevents a full thermalization. The intrinsic reset effect of environmental noise is exploited to increase the ground state population when two spins reach the distant configuration. One can conclude that, even in the presence of a very hot environment, there is still a chance to see entanglement in an oscillating molecule after an arbitrarily long time; see figure 5.

### 3.3. Beat dephasing via thermalization

The constructive role played by the environmental noise can be emphasized by including an additional source of decoherence to further destroy entanglement. Let us consider a ‘phenomenological’ pure dephasing process acting on the two spins via the term

$$D_p \rho(t) = \gamma_p \sum_{i=1}^{2} \left[ \sigma_z^{(i)} \rho(t) \sigma_z^{(i)} - \rho(t) \right],$$

with $\gamma_p$ denoting the dephasing rate. As is well known, pure dephasing destroys all the coherence and always opposes the generation of entanglement. Even for the oscillating molecule, the effect of pure dephasing alone is to remove the off-diagonal density matrix elements of the two-spin state, and thereby destroy entanglement in the long run. This is evident in figure 7, in which entanglement can be found only in the first oscillation period and certainly not in the asymptotic cycle (i.e. for $t \to \infty$). What happens in the presence of a hot thermal bath? One would expect that entanglement were destroyed faster due to this extra decoherence source, and this is indeed true as long as only the first oscillation period is taken into account; see figure 7. However, the
Figure 7. Entanglement $C$ (a) and ground state population $p_0$ (b) versus the molecular configuration $d$. The dephasing rate is $\gamma_0 = 0.02$. The system–bath coupling strength is $\kappa = 0$ (no bath) or $\kappa = 0.01$. The parameters are the same as for figure 5(a).

The constructive effect of thermalization becomes evident in the successive cycles in which it partly counteracts the pure dephasing through its built-in reset mechanism of re-pumping the ground state population (as explained in the above subsection).

3.4. Stochastic classical motion

So far, we have considered a deterministic and purely periodic classical motion. In the context of biological systems, a more realistic approach naturally includes a certain degree of stochasticity. Different scenarios are conceivable depending on the biological situations that one intends to model.

3.4.1. Fluctuating configurations. The allosteric process depicted in figure 1 is usually triggered by the interaction with a second chemical compound: when the molecule is in the open configuration, a chemical docks on the molecule and causes its allosteric contraction. The molecule reaches its closed configuration and stays there until the chemical is released. When the chemical undocks, the molecule opens [29, 34].

The waiting times in the open and the closed configuration can be thought of as two stochastic variables which are, in principle, different. Supposing normal (Gaussian) but independent distributions for both processes, one obtains trajectories like the one depicted in figure 8, where the linear change in the spin distance during the conformational process is an arbitrary but nonessential choice.

Owing to our understanding obtained in the deterministic case, the intuition correctly predicts the limiting cases of long and short waiting times: when the chemical frequently docks or undocks from the molecule, the system is continuously kept out of equilibrium and its dynamics closely resembles the deterministic case. On the other hand, for long waiting times (i.e. long compared to the thermalization rate) equilibrium is reached in the extreme configurations, and during the contraction of the molecule the single-shot behavior is reproduced. The creation of entanglement is here only a transient effect.
Figure 8. Example of a trajectory for waiting times distributed according to normal distributions (neglecting their tails for negative values). The mean $\bar{d}$ and standard deviation $\sigma$ in the open (closed) configuration are $\bar{d} = 14.5$, $\sigma = 6$ ($\bar{d} = 7.5$, $\sigma = 5$), respectively.

Figure 9. Energy landscape $V(d)$ of molecular configurations with respect to the spin distance $d$. For consistency, we also set the temperature of this environment to $\beta = 1$ and choose the barrier height accordingly. Explicitly, the potential is described by $V(d) = a_v(d - 30)^2 + b_v(d - 30)^4 + h_v$, with $b_v = 0.0004$, $a_v = -128 b_v$ and $h = a_v^2/(4b_v)$.

3.4.2. Potential landscape and thermal vibrations. A somewhat more interesting dynamics is obtained in the following scenario: the molecule has two configurations which minimize the potential energy associated with its classical shape. In our model, it is immediate to identify the distance between the spins with an effective collective position coordinate for the macromolecule: varying such a coordinate we move from the low-energy closed configuration to the low-energy open configuration via an energy landscape that contains barriers. For the sake of simplicity, we model such a energy surface as a double-well potential presenting a single central barrier and described by the quartic potential shown in figure 9.
Once a configuration is chosen (as an arbitrary initial condition), the molecule starts exploring the other possible configurations due to the thermal energy supplied by the surrounding environment (note that it is a totally different and unrelated environment with respect to the bosonic heat bath). It means that the molecule can randomly contract or stretch, as long as the energy of the new configuration is within thermal fluctuations. Using the Metropolis algorithm \[35\] and the potential landscape of figure 9, we obtain trajectories such as the one shown in figure 10.

In this situation, one cannot describe the molecular motion as composed of distinct opening and closing processes. In fact, the molecule stretches and contracts its configuration only partially and in a stochastic way. Nevertheless, the mechanisms presented in section 3.2 are still at work in this scenario. Due to the increasing energy gap, the ground state population rises when the distance between the spins increases, with an ‘inertial’ reaction time determined by the thermalization rate. Whenever the system undergoes a relatively fast contraction after having spent enough time in an extended configuration, entanglement is generated, as is clearly seen in figure 11.

3.5. The non-Markovian case

The microscopic derivation of the Lindblad-type master equation previously introduced in this section relies on several approximations (for a more detailed derivation see \[28\]). These approximations refer to assumptions regarding the time scale of the thermal bath’s dynamics and the strength of its interaction with the system. In particular, such an interaction is supposed to be weak and the bath’s relaxation time very short compared with the time scale of the system dynamics. For many biological processes, these conditions may not be satisfied. Nevertheless, they can be considered as a useful working hypothesis to obtain analytical expressions (suitable for numerical integration) and to illustrate our main idea.

To demonstrate that such simplifying assumptions are indeed not essential for the validity of our arguments, we now extend our analysis to situations with a not-so-weak system–bath interaction and a non-Markovian reduced dynamics. We rephrase our problem within the path integral formulation of quantum mechanics: in this formulation, the actual evolution of a
quantum state is determined by the coherent sum of all possible evolutions weighted via a phase proportional to the corresponding (classical) action. The presence of an environment in contact with the system leads to an additional weighting factor, the so-called influence functional [36, 37]. The influence functional depends on the system’s possible evolution and on the properties of the bath, and in general is nonlocal in time. This formalism is particularly convenient in the case of environments composed of noninteracting harmonic oscillators, where one can obtain an analytical expression for the influence functional [37].

The part of the summation involving the system’s degrees of freedom has to be performed numerically, and a particularly favorable prescription was introduced by Makri and Makarov under the acronym of QUAPI (QUasi Adiabatic propagator Path Integral) [38, 39]. Their method allows one to include a finite-time memory via a truncated influence functional that takes into account only a few of the last propagation steps. The memory time is determined by how many time steps are included and by how long each of them lasts: $T_{\text{mem}} = \Delta k \Delta t$. In principle, the longer the $T_{\text{mem}}$ the further one can relax the Markovian assumption. In practice, however, the computational effort scales exponentially with $\Delta k$ and the error in the single-step propagation quadratically in $\Delta t$. To overcome these limitations, each simulation has to be performed with different parameters and one has to perform two extrapolation procedures: the first keeps the $T_{\text{mem}}$ constant while $\Delta t \to 0$ to obtain a precise estimate with finite memory, the second increases $T_{\text{mem}} \to \infty$ to extrapolate the infinite memory time limit.

These technical procedures are extensively presented in [40] but are beyond the scope of this work: here we do not want to precisely quantify the amount of entanglement generated under a specific system–environment interaction, but our goal is to demonstrate that the mechanism we introduced is indeed robust and is not confined to the Born–Markov regime. However, we have selectively performed the extrapolation procedures for several points along the curves: the agreement is always good (at the percentage level) and consistent with the values of entanglement found before the extrapolations. The extent of the memory effects is determined by the so-called bath response function BRF; to properly take into account the non-Markovianity of the environment, the memory time $T_{\text{mem}}$ has to be larger than the bath correlation time given by the width of the BRF; see figure 12. Figure 13 shows how the generation of entanglement persists even for baths with finite-time correlations (determined by

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**Figure 11.** Ground state (upper, blue) and concurrence (lower, red) in the presence of the thermally driven fluctuations of the molecule’s configuration.
a small value of the cut-off frequency in the spectral density $J(\omega)$, giving rise to finite memory effects) and moderately strong system–environment couplings. In fact, it has been shown that non-Markovian environments can even help in sustaining quantum coherence and entanglement in related systems [15, 41].

4. The spin gas model

The actual environment for biological systems is expected to be very specific and complex and has not yet been fully characterized. In the above section, we investigated the case of thermal baths composed of a large number of harmonic oscillators, but this is most probably still a simplistic (even if reasonable and common) choice. In this perspective, it is worth seeing whether the main features we observe for bosonic heat baths are preserved also for a different kind of environment, namely the spin gas model [42–44], which is particularly suggestive in a biological context. Here the environment is composed of particles that follow random classical trajectories and collide with our two spins. During the collisions they stochastically interact with the spins, giving rise to an effective decoherence process. Under certain assumptions [44], at a coarse time level that includes both the coherent Hamiltonian dynamics and the random collisions, it is possible to derive a phenomenological description in terms of a Lindblad-type quantum master equation characterized by the following dissipator:

$$\mathcal{D}\rho(t) = \gamma \sum_{\alpha=1}^{2} [\mathcal{L}^{(\alpha)}_{g} \rho(t) + \mathcal{L}^{(\alpha)}_{d} \rho(t)],$$

with

$$\mathcal{L}^{(\alpha)}_{g} \rho = s[2\sigma_{+}^{(\alpha)} \rho \sigma_{-}^{(\alpha)} - \rho \sigma_{+}^{(\alpha)} \sigma_{+}^{(\alpha)} - \sigma_{-}^{(\alpha)} \sigma_{+}^{(\alpha)} \rho],$$

$$\mathcal{L}^{(\alpha)}_{d} \rho = (1-s)[2\sigma_{-}^{(\alpha)} \rho \sigma_{-}^{(\alpha)} - \rho \sigma_{-}^{(\alpha)} \sigma_{-}^{(\alpha)} - \sigma_{+}^{(\alpha)} \sigma_{-}^{(\alpha)} \rho],$$

Figure 12. The bath response function for the Markovian ($\omega_c = 40$, (a)) and non-Markovian ($\omega_c = 4$, (b)) regimes. The red solid curve is the real part of the BRF, the green dashed curve is its imaginary part and the black vertical lines mark the memory time $T_{mem} = 1.5$ considered in figure 13.
where $\sigma^{(a)}_{\pm} = (\sigma^{(a)}_{x} \pm i \sigma^{(a)}_{y})/2$ are the ladder operators for a two-level system. The decoherent channels $L^{(i)}_g(\rho)$, $L^{(i)}_d(\rho)$ represent the energy gain and loss during the process: $\gamma$ is the rate at which $\langle (1 + \sigma_z)/2 \rangle$ decays, and $s$ is its equilibrium value [45]. Without loss of generality, we assume that $\gamma > 0$ and $0 \leq s \leq 1/2$. We also note that correlations in the spin gas can be included in the analysis [46].

4.1. Static entanglement in the steady state

Similar to the case of the bosonic heat baths, we first consider the steady state $\rho \equiv \rho(t \to \infty)$ for each molecular configuration, i.e. at fixed spin–spin interaction strength $J$ and background field $B$, to verify that it is a separable state. To this end, we impose that

$$-i[H_M(t), \rho] + \mathcal{D}\rho = 0.$$  

Figure 13. The entanglement $C$ (a, c) and ground state population $p_0$ (b, d) versus molecular configuration $d$ for a non-Markovian environment. The blue dot-dashed curve indicates the first period; the red solid curve marks the asymptotic cycle, while the green dotted curve refers to the QUAPI equilibrium state for each specific configuration. Top: $\omega_c = 4$, $\kappa = 0.01$, $T_{\text{mem}} = 1.5$, $\tau = 100$, $\Delta t = 5$; bottom: $\omega_c = 4$, $\kappa = 0.05$, $T_{\text{mem}} = 1.5$, $\tau = 75$, $\Delta t = 5$, while the other parameters are as in figure 5(a).
Figure 14. The critical value of $s$ (above which no static entanglement exists in the steady state) for different values of $B$ and $J$ with $\gamma = 0.025$.

It is straightforward to check that the steady state for each configuration $d$ is unique and that its nonzero density matrix elements are given by

\[
\rho_{00} = \frac{[J^2 + 4b^2s^2]}{4(J^2 + b^2)}, \\
\rho_{11} = \frac{[J^2 + 4b^2s(1-s)]}{4(J^2 + b^2)}, \\
\rho_{22} = \frac{[J^2 + 4b^2s(1-s)]}{4(J^2 + b^2)}, \\
\rho_{33} = \frac{[J^2 + 4b^2(1-s)^2]}{4(J^2 + b^2)}, \\
\rho_{03} = -J(2B - i\gamma)(1 - 2s)/2(J^2 + b^2), \\
\rho_{30} = -J(2B + i\gamma)(1 - 2s)/2(J^2 + b^2),
\]

(29)

where $b = (4B^2 + \gamma^2)^{1/2}$. The static entanglement of the steady state quantified by concurrence is calculated as

\[
C(\rho) = 2 \max\{0, |\rho_{03}| - (\rho_{11}\rho_{22})^{1/2}\},
\]

resulting in

\[
C(\rho) = 2 \max 0, \frac{2Jb(1 - 2s) - [J^2 + 4b^2(1 - s)s]}{4(J^2 + b^2)}
\]

(30)

Thus, no static entanglement is possible if the parameter $s$ exceeds the critical value of

\[
s_c = \max \left\{ 0, \frac{1}{2} \left[ 1 + J/b - (1 + 2J^2/b^2)^{1/2} \right] \right\}
\]

(31)

plotted in figure 14.

4.2. Persistent dynamic entanglement

The physical picture underlying the spin gas model is very different from the collection of harmonic oscillators constituting the bosonic thermal bath and these two environments destroy entanglement according to different mechanisms. Nevertheless, they share the essential features that allow cyclic generation of fresh entanglement via the constructive role played by the environment noise itself. Even when in any possible static configuration the steady states for both models and high enough temperature (bosonic heat baths) or level of noise (the spin gas environment with a large value of $s$) do not exhibit entanglement, entanglement persistently appears when classical oscillations are taken into account.
Figure 15. The entanglement $C$ (a) and ground state population $p_0$ (b) versus the molecular configuration $d$ for the spin gas environment. The blue dot-dashed curve indicates the first period; the red solid curve marks the asymptotic cycle; the green dotted curve refers to the steady state for a specific configuration. The decoherence rates are $s = 0.16$ and $\gamma = 0.025$; the oscillation parameters are the same as in figure 5(a).

Figure 16. The entanglement $C$ (a) and ground state population $p_0$ (b) versus the molecular configuration $d$ for the spin gas environment with additional pure dephasing. The blue dot-dashed curve indicates the first period; the red solid curve marks the asymptotic cycle; the green dotted curve refers to the steady state for a specific configuration. The decoherence rates are $s = 0.16$, $\gamma = 0.025$ and $\gamma_p = 0.01$; the oscillation parameters are the same as in figure 5(a).

This can be explained since the spin gas environment also contains a built-in reset mechanism via spin flip: for the molecular configuration in which the two spins are spatially separated, the environment effectively lowers the system’s entropy and increases the ground state population. Thus, under these conditions and in accordance with what has been observed for thermal baths, dynamic entanglement is generated in the long time limit (figure 15).

As in the case of the bosonic heat baths, here the constructive role of the environmental noise can also be further illustrated by adding pure dephasing. One can see from figure 16...
that the noise in the spin gas model can counteract the effect of pure dephasing and sustain the persistent dynamic entanglement. The green curve is here obtained from an expression analogous to equation (29) and generalized to include the additional dephasing.

5. Concluding remarks

In this work, we have demonstrated how entanglement can persist as a recurrent property even in systems exposed to a hot and noisy environment. To achieve this task we have exploited the presence of an internal driving force which keeps the system out of the corresponding equilibrium state. In a biologically inspired context, such a driving force can be identified with conformational changes, including the stretching of macromolecules or their folding/unfolding processes. Here, we describe these conformational changes as purely classical, but capable of influencing the dynamics of localized quantum degrees of freedom and of preventing complete thermalization. The nonequilibrium interplay between classical motion and open quantum system dynamics gives rise to an effective reset mechanism which promotes the generation of fresh entanglement even for arbitrary long times. The present model serves two purposes: on the one hand, it provides a concrete route where to search for signatures of entanglement in biomolecular systems operating at room temperature. On the other hand, it indicates new directions to design molecular scale machines able to generate nontrivial quantum states even in noisy environments. Finally, let us remark that an experimental simulation of our model is achievable with state-of-the-art technology in setups such as ion traps or nano-mechanical systems.

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