Forced synchronization of a quantum dissipative dynamics

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Abstract. We generalize the phenomenon of forced stochastic synchronization into the quantum domain within the framework of a paradigmatic spin-boson model (tunneling charge, or flipping spin 1/2 coupled to an environment) which is driven by an external periodic rectangular field. The overdamped regime of dissipative quantum tunneling is studied. Thermal noise assisted synchronization of a very high quality is shown to occur in a broad range of temperatures, driving strengths and frequencies, if the external driving frequency exceeds the zero-temperature limit of dissipative tunneling rate, the dissipation strength exceeds a critical value, and the driving is sufficiently strong. A simple criterion for such stochastic synchronization is established. Both the similarities and the profound differences with the akin phenomenon of quantum stochastic resonance are outlined.

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INTRODUCTION

Synchronization is a very universal phenomenon in *classical physics* [1]. In particular, forced synchronization refers to the locking of the phase of a nonlinear driven oscillator to that of a periodic driving field. In the presence of noise, it is also possible to define a stochastic clock with some mean phase frequency which depends on the noise strength. The ordering and locking of this stochastic clock to the pacemaker driving clock within some noise range is called stochastic synchronization [2]. The less probable phase slips between the driver and the driven system are, the better is the quality of synchronization. Can a *quantum stochastic clock* provided by a tunneling charge jumping at random times between two different sites of localization in a dissipative environment, or a quantum spin 1/2 flipping at random between two orientations be synchronized with a periodic field and under what conditions? We answer this intriguing question within the framework of the paradigmatic spin-boson model in the presence of an external rectangular driving.

MODEL, THEORY, AND RESULTS

The model is described by the following Hamiltonian

$$\hat{H}(t) = \frac{1}{2}\varepsilon(t)\hat{\sigma}_z + \frac{1}{2}\hbar\Delta\hat{\sigma}_x + \frac{1}{2}\hat{\sigma}_z\sum_j\kappa_j(\hat{b}_j^{\dagger} + \hat{b}_j) + \sum_j\hbar\omega_j(\hat{b}_j^{\dagger}\hat{b}_j + \frac{1}{2}),$$
(1)

wherein, the operators $\hat{\sigma}_z$ and $\hat{\sigma}_x$ denote the standard Pauli operators, $\varepsilon(t)$ is a timedependent energy bias, and $\hbar\Delta$ is the tunnel matrix element. The bath Hamiltonian [last term in Eq. (1)] is expressed in terms of the operators \hat{b}_j^+ and \hat{b}_j associated to the *j*th bath normal mode with frequency ω_j . The stochastic influence of the quantum thermal bath is captured by an *operator* random force $\hat{\xi}(t) = \sum_j \kappa_j (\hat{b}_j^{\dagger} e^{i\omega_j t} + \hat{b}_j e^{-i\omega_j t})$. It can be characterized by the spectral density $J(\omega) = (\pi/\hbar) \sum_j \kappa_j^2 \delta(\omega - \omega_j)$. We assume that $J(\omega)$ acquires the Ohmic form, $J(\omega) = 2\pi\hbar\alpha\omega e^{-\omega/\omega_c}$, with friction strength α and an exponential cutoff. We consider the overdamped limit $\alpha > 1/2$ and weak tunneling limit $\Delta \ll \omega_c$. Then, the dynamics of the diagonal elements of the reduced density matrix, $p_{\beta}(t) = p_{\pm}(t)$, is given by the Pauli master equation [3]

$$\dot{p}_{\beta}(t) = W_{-\beta}(t)p_{-\beta}(t) - W_{\beta}(t)p_{\beta}(t) , \qquad (2)$$

where the time-dependent relaxation rates $W_{\beta}(t)$ within the Golden Rule approximation and the approximation of adiabatically slow varying bias $\varepsilon(t)$ read:

$$W_{\pm}(t) = \frac{1}{2}\Delta^2 \int_0^\infty d\tau \exp[-Q'(\tau)] \cos\left[Q''(\tau) \mp \frac{1}{\hbar}\varepsilon(t)\tau\right] \,. \tag{3}$$

The functions Q'(t) and Q''(t) denote the real and imaginary parts of the dissipation kernel

$$Q(t) = \frac{i\lambda t}{\hbar} + \frac{1}{\hbar^2} \int_0^t dt_1 \int_0^{t_1} dt_2 \langle \hat{\xi}(t_2) \hat{\xi}(0) \rangle_T , \qquad (4)$$

wherein $\lambda = \int_0^\infty d\omega J(\omega)/(\pi\omega)$ is the bath reorganization energy [3]. For the considered model one finds that $\lambda = 2\alpha\hbar\omega_c$ and

$$Q'(t) = 2\alpha \ln \left\{ \sqrt{1 + \omega_c^2 t^2} \frac{\Gamma^2(1+\kappa)}{|\Gamma(1+\kappa+i\omega_T t)|^2} \right\}$$
(5)

$$Q''(t) = 2\alpha \arctan(\omega_c t).$$
(6)

In Eq. (5), $\Gamma(z)$ denotes the Gamma function, $\omega_T = k_B T/\hbar$, and $\kappa = \omega_T/\omega_c$. The quantum rates satisfy $W_-(t) = \exp(-\varepsilon(t)/(k_B T))W_+(t)$ for each instant of time *t*. At zero temperature,

$$W_{\pm,T=0}(t) = \boldsymbol{\theta}[\pm \boldsymbol{\varepsilon}(t)] \frac{\pi \Delta^2}{2\omega_c \Gamma(2\alpha)} \left(\frac{\pm \boldsymbol{\varepsilon}(t)}{\hbar \omega_c}\right)^{2\alpha - 1} \exp[\mp \boldsymbol{\varepsilon}(t)/(\hbar \omega_c)],$$

where $\theta(t)$ is the Heaviside step function [4], i.e. one of the rates is nonzero, while another one is exactly zero. In the limit of high-temperatures (classical environment) and quasi-static environmental noise limit, the rates are well approximated by the celebrated Marcus-Levich-Dogonadze rate expression $W_{\pm}(t) = (\pi/2)\hbar\Delta^2/\sqrt{4\pi\lambda k_B T} \exp[-(\pm\varepsilon(t) - \lambda)^2/(4\lambda k_B T)]$. In a fully quantum regime for $k_B T < \hbar\omega_c$, the rates have to be evaluated numerically.

Our thought-experimental setup mimics the quantum analogue of a classical (phase)synchronization behavior elaborated in Refs. [5, 6]. A quantum particle with the charge q (electron, or proton) makes tunneling (instant) jumps forth and back between two sites of localization separated by the distance r_0 . The energy bias is modulated in time by the applied electric field $\mathscr{E}(t)$ yielding $\varepsilon(t) = qr_0\mathscr{E}(t)$ (alternatively, it could be a spin 1/2 and a magnetic field). The external field alternates also dichotomously, changing its direction, however, periodically in time. The tunneling jump process can be described mathematically as a classical telegraph noise defined by the master equation (2) with rates which are fully quantum-mechanical and time-dependent.

We are interested in the synchronization of tunneling events separated by random time intervals with the external drive alternations. One then counts the number of jumps n(t) within a time window $[t_0,t)$. Following Ref. [5], we introduce the random phase $\phi(t,t_0) = \pi n(t)$, which increases by π at each switching event (two subsequent switches correspond to a 2π -cycle of random duration), and define the average frequency and diffusion coefficients associated to the stochastic phase-process as $\overline{\Omega}_{ph} := \lim_{t\to\infty} \langle \phi(t,t_0) \rangle / (t-t_0)$ and $2\overline{D}_{ph} := \lim_{t\to\infty} \left[\langle \phi^2(t,t_0) \rangle - \langle \phi(t,t_0) \rangle^2 \right] / (t-t_0)$, respectively. Using a stochastic path-integral description of the driven telegraph process [7], the following exact results were obtained for a periodic rectangular driving $\varepsilon(t) = \pm \varepsilon_0$ with amplitude ε_0 and frequency $\Omega = 2\pi/\mathcal{T}$ [6, 8]

$$\overline{\Omega}_{\rm ph} = \frac{\pi W}{2} \left\{ 1 - \delta p_{\rm eq}^2 \left[1 - \frac{4 \tanh \left(W \mathscr{T} / 4 \right)}{W \mathscr{T}} \right] \right\},\tag{7}$$

and

$$2\overline{D}_{ph} = \pi \overline{\Omega}_{ph} - \frac{2\pi^2}{\mathscr{T}} \delta p_{eq}^4 \tanh^3 (W \mathscr{T}/4) - \frac{\pi^2}{2\mathscr{T}} \delta p_{eq}^2 \left(1 - \delta p_{eq}^2\right) \left\{ 12 \tanh (W \mathscr{T}/4) - W \mathscr{T} \left[1 + 2 \operatorname{sech}^2 (W \mathscr{T}/4)\right] \right\}.$$
(8)

Here, W denotes the sum of the forward and backward rates in Eq. (3) for a fixed value of the field \mathscr{E}_0 , i.e., for $\varepsilon(t) = \varepsilon_0 = qr_0\mathscr{E}_0$, and $\delta p_{eq} = \tanh(\varepsilon_0/(2k_BT))$ is the absolute value of the difference of the equilibrium populations. The inverse Fano factor of the counting process $R := \pi \overline{\Omega}_{ph}/(2\overline{D}_{ph})$ provides a reliable quality measure of forced synchronization [2, 5]. It corresponds to the number of tunneling jumps synchronized to the driving alternations. Desynchronization occurs, when the variance of the synchronized counting process becomes about unity, $\langle \overline{\delta n^2}(t_{synchro}) \rangle = 1$ (a phase slip occurs). From this condition, the (de)synchronization time follows as

$$t_{\rm synchro} \sim R \mathscr{T}/2$$
.

Criterion for stochastic synchronization

For stochastic synchronization to occur the particle must have sufficient time to make a transition to the lower energy state during the tilting half-period. This requires that the mean residence time $\langle \tau_+(\varepsilon) \rangle = 1/W_+(\varepsilon_0)$ in the higher energy state should be much less than $\mathscr{T}/2$. On the other hand, the backward transitions during such a forward tilt must be prohibited. The synchronization criterion thus reads,

$$1/W_{+}(\varepsilon_{0}) \ll \mathscr{T}/2 \ll 1/W_{-}(\varepsilon_{0}) = \exp(\varepsilon_{0}/k_{B}T)/W_{+}(\varepsilon_{0}) .$$
(9)

It should be contrasted with the criterion for Stochastic Resonance (SR) reading [9]

$$1/W_{+}(\varepsilon_{0}) = \mathscr{T}/2 \text{ or } \pi W_{+}(\varepsilon_{0}) = \Omega.$$
 (10)

Clearly, for a weak driving, within the linear response (LR) approximation, $\varepsilon_0 \ll k_B T$, the criterion (9) can never be justified, while SR can occur [9]. Moreover, SR is frequently not related to synchronization at all. In particular, the earlier works on quantum SR (QSR) [10] within the same spin-boson model uncovered, within the parameter regime $\alpha < 1$, that QSR in the considered symmetric (in the absence of driving) system is impossible, and an additional static bias is required [9]. This even was thought to be a main feature of QSR, as compared with the classical SR [9]. Such QSR in a biased spinboson system has, however, no relation to synchronization, as it does not correspond to any matching of the time scale of driving and that of the dissipative tunneling dynamics. In [11] it was shown, however, that QSR is also possible within the LR theory for the studied symmetric model provided that quantum friction is sufficiently strong, $\alpha > 1$. This parameter regime is relevant, e.g. for nonadiabatic electron tunneling in condensed molecular systems [3], where α can be as large as $\alpha \sim 5 - 10$ and even larger.

We were guided by this earlier QSR research to find the parameter regime, where the thermal noise-assisted *Quantum Stochastic Synchronization* (QSS) can occur. A fundamental difference of QSS, as compared with its classical counterpart, is that QSS is expected to occur always at T = 0, if the driving is sufficiently slow, $\Omega \ll W_{T=0}(\varepsilon_0)$. This is so because one of the rates is exactly zero, as it follows from the detailed balance condition. Thus, a desynchronization transition occurs with enhancing the temperature above some threshold value [8]. The question is, however, whether the thermal noise can help to synchronize when $\Omega > W_{T=0}(\varepsilon_0)$ and the particle cannot follow the bias alternations at T = 0. As in the case of QSR, this question is highly nontrivial because of the non-Arrhenius, power law temperature dependence of the tunneling rates. Our numerical study based on the outlined analytical theory revealed that such a quantum thermal noise assisted QSS is only possible when $\alpha > 1$, in agreement with the QSR research. Moreover, large $\alpha \sim 5 - 10$ are preferable.

Numerical results

Let us first illustrate a fundamental difference between QSR and QSS for a large friction $\alpha = 10$, some fixed tunnel matrix element $\Delta = 4 \cdot 10^{-4}$, frequency $\Omega = 10^{-11}$ and

for a moderate driving strength $\varepsilon_0 = 0.5$ which is already beyond the LR condition. All the dimensional quantities are scaled ($\hbar = 1, k_B = 1$) in terms of the quantum crossover temperature $T_c = \hbar \omega_c / k_B$ of thermal bath oscillators. The parameters are chosen to mimic nonadiabatic electron tunneling in the molecular dimers of azurin [8].



FIGURE 1. (a) Mean stochastic frequency $\overline{\Omega}_{ph}$, mean phase diffusion coefficient \overline{D}_{ph} and (b) the inverse Fano factor *R* versus the scaled temperature κ for the driving strength $\varepsilon_0 = 0.5$. Other parameters are given in the text.

Fig. 1(a) shows that synchronization for the chosen parameters is absent. The mean phase frequency $\overline{\Omega}_{ph}$ as a function of the scaled temperature κ crosses the line $\Omega = 10^{-11}$ at a certain point. However, any frequency locking supplemented by a minimum of the phase diffusion coefficient is absent. Therefore, this is not a synchronization, per definition, as any synchronization requires a frequency locking. The crossing point around $\kappa \approx 0.5$ corresponds, however, to QSR as it can be deduced from Fig. 1(b). Indeed, the quality, or coherence factor *R* displays a smooth maximum when quantum stochastic resonance occurs. The amplitude of this maximum is, however, so small that desynchronizing phase slips occur permanently, even where a driving-induced phase coherence, R > 1, exists. Such QSR can be considered also as a kind of coherence resonance [12] induced by the external driving. It constitutes a precursor of quantum stochastic synchronization that occurs with a further increase of the driving strength above some threshold $\varepsilon_0 \approx 2.5$, for other parameters fixed. For $\varepsilon_0 = 5$ the quality of synchronization is already very high as evidenced by Fig. 2.



FIGURE 2. The same as Fig. 1 for the driving strength $\varepsilon_0 = 5$.

In this figure, the mean phase frequency $\overline{\Omega}_{ph}$ is clearly locked to the external frequency Ω in a broad range of temperatures, while the mean phase diffusion coefficient \overline{D}_{ph} displays a pronounced minimum at some temperature. At this minimum, the quality of the synchronization is astonishingly high [see Fig. 2(b)] reaching the maximum about 10⁵ phase-locked tunneling jumps before a phase slip occurs.

Our estimations for electron tunneling in molecular dimers like azurin show [8] that for tunneling distances $r_0 \sim 15$ Å, crossover temperatures $T_c \sim 150$ K, tunnel couplings $\hbar\Delta \sim 5 \cdot 10^{-3}$ meV, the required driving frequencies are in the range from several Hz to several hundreds Hz, while the electrical field strength should be about $5 \cdot 10^4$ V/cm to arrive at a high quality synchronization. Such experimental studies should be feasible in laboratories.

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