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Periodic thermodynamics of a two spin Rabi model

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Abstract. We consider two $s = 1/2$ spins with Heisenberg coupling and a monochromatic, circularly polarized magnetic field acting only onto one of the two spins. This system turns out to be analytically solvable. Also the statistical distribution of the work performed by the driving forces during one period can be obtained in closed form and the Jarzynski equation can be checked. The mean value of this work, viewed as a function of the physical parameters, exhibits features that can be related to some kind of Rabi oscillation. Moreover, when coupled to a heat bath the two spin system will approach a non-equilibrium steady state (NESS) that can be calculated in the golden rule approximation. The occupation probabilities of the NESS are shown not to be of Boltzmann type, with the exception of a single phase with infinite quasitemperature. The parameter space of the two spin Rabi model can be decomposed into eight phase domains such that the NESS probabilities possess discontinuous derivatives at the phase boundaries. The latter property is shown to hold also for more general periodically driven N-level systems.

Keywords: rigorous results in statistical mechanics, thermalization

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Contents

1. Introduction

A quantum system developing according to a time-dependent Hamiltonian $H(t)$ which varies periodically with time t, such that

$$H(t) = H(t + T) \; , \tag{1}$$

possesses a complete set of *Floquet states*, that is, of solutions to the time-dependent Schrödinger equation having the particular form

$$\psi_n(t) = u_n(t) \, \exp(-\mathrm{i}\varepsilon_n t) \; . \tag{2}$$

The *Floquet functions* $u_n(t)$ are also T-periodic and the quantities ε_n are known as *quasienergies* [1–3]. They are only uniquely determined up to integer multiples of the driving frequency $\omega = \frac{2\pi}{T}$.

The significance of these Floquet states (2) is based on the fact that every solution $\psi(t)$ to the time-dependent Schrödinger equation can be expanded with respect to the Floquet basis,

$$\psi(t) = \sum_n c_n \, u_n(t) \, \exp(-\mathrm{i}\varepsilon_n t) \; , \tag{3}$$

such that the coefficients c_n do not depend on time. Hence, the Floquet states propagate with constant occupation probabilities $|c_n|^2$, despite the presence of a time-periodic drive. However, if the periodically driven system is interacting with an environment, as it happens in many cases of experimental interest [4–9], that environment may continuously induce transitions among the system's Floquet states. This has the effect that after some relaxation time a quasi-stationary distribution $\{p_n\}$ of Floquet-state occupation probabilities is reached which contains no memory of the initial state. The question arises how to quantify this distribution.

In a short programmatic note entitled 'Periodic Thermodynamics', Kohn [10] has drawn attention to such quasi-stationary Floquet-state distributions $\{p_n\}$. In an earlier pioneering study, Breuer *et al* had already calculated these distributions for time-periodically forced oscillators coupled to a thermal oscillator bath [11]. To date, a great variety of different individual aspects of the 'periodic thermodynamics' envisioned by Kohn has been discussed in the literature [12–24], but a coherent overall picture is still lacking.

In this situation it seems advisable to resort to models which are sufficiently simple to admit analytical solutions. Recent results into this direction are the following:

- As mentioned above, for the particular case of a linearly forced harmonic oscillator the authors of [11] have shown that the Floquet-state distribution remains a Boltzmann distribution with the temperature of the heat bath, see also [25].
- Similarly, the parametrically driven harmonic oscillator assumes a quasi-stationary state with a quasi-temperature that is, however, generally different from the bath temperature, see [26, 27].
- A spin s exposed to both a static magnetic field and an oscillating, circularly polarized magnetic field applied perpendicular to the static one, as in the classic Rabi set-up [28], and coupled to a thermal bath of harmonic oscillators has been shown to approach a quasi Boltzmann distribution, see [29]. This work generalizes the results of [25] for the case $s = 1/2$.

In the present work we will consider, similarly as in [25], an $s = 1/2$ spin with a circularly polarized driving but only coupled to the heat bath via another $s = 1/2$ spin, see figure 1. An analogous system has previously been numerically investigated with the focus on decoherence [30]. In order to keep the analytical treatment as simple as possible we will set $\omega = \omega_0 = 1$, where ω_0 denotes the dimensionless Larmor frequency of the static magnetic field. Then it is possible to explicitly calculate the quasienergies ϵ_n and the probabilities p_n, $n = 1, \ldots, 4$ of the NESS, although the latter are too complex to be given in closed form. It turns out that the p_n are *not* of Boltzmann type thereby rigorously confirming the general conjectures about the nature of the NESS for a simple system. Another result will be the partition of the parameter space $\boldsymbol{\mathcal{P}}$ into certain *phases* $\boldsymbol{\mathcal{P}}_\nu$ such that the p_n, while being smooth functions of the parameters within the phases $\boldsymbol{\mathcal{P}}_\nu$, will have discontinuous derivatives at the phase boundaries. These findings will also hold for general periodically driven N-level systems. For the special system under consideration we additionally observe that all four NESS probabilities coincide for a certain phase A which could be formally understood as an infinite quasitemperature of this phase. But we will provide arguments that this result is confined to this very system and will probably not hold in general.

The paper is organized as follows. In section 2 we define the system to be studied and derive its explicit time evolution in the Floquet normal form. The time evolution matrix for one period (monodromy matrix) of the present system turns out to be symmetric and hence possesses real eigenvectors. The proof of this has been moved

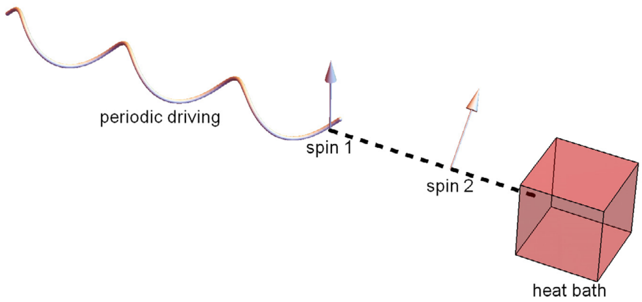

Figure 1. Schematic representation of the two spin Rabi model considered in this paper.

to an appendix A. The explicit results on the time evolution are used in section 3 to calculate the statistical distribution of the work performed by the periodic driving during one period and to check our results by confirming the corresponding Jarzynski equation As a by-product we prove the physically plausible fact that the expectation value of the work is always non-negative and discuss the mean value of the work. The general golden-rule approach to periodic thermodynamics is briefly recapitulated in section 4.1 and applied to the two spin system under consideration in section 4.2. The partition of the parameter space into phases and the 2^{nd} order phase transitions at the phase boundaries seems to hold also for the general case of periodically driven N-level systems. The pertinent arguments are presented in the appendix B. We close with a summary and outlook in section 5.

2. Definitions and general results

We consider two spins with $s = 1/2$ and the composite system described by the four-dimensional Hilbert space $\mathcal{H} = \mathbb{C}^2 \otimes \mathbb{C}^2$. The static Hamiltonian is assumed to be of the form

$$H_0 = \underset{\sim}{\mathbf{s}}_3^{(1)} \otimes \mathbb{1} + \mathbb{1} \otimes \underset{\sim}{\mathbf{s}}_3^{(2)} + \lambda\, \underset{\sim}{\mathbf{s}}^{(1)} \cdot \underset{\sim}{\mathbf{s}}^{(2)}, \tag{4}$$

where $\underset{\sim}{\mathbf{s}}^{(1)}$ and $\underset{\sim}{\mathbf{s}}^{(2)}$ are the usual $s = 1/2$ vector spin operators for the subsystems and $\lambda > 0$ is some coupling parameter. The eigenvalues E_n of H_0 are

$$E_{1,2} = \frac{\lambda}{4} \pm 1,\; E_3 = -\frac{3\lambda}{4},\; E_4 = \frac{\lambda}{4}. \tag{5}$$

The periodic circularly polarized driving with amplitude f and unit angular frequency acts only on the first spin and thus the total Hamiltonian can be written as

$$H(t) = H_0 + f\left(\cos t\, \underset{\sim}{\mathbf{s}}_1^{(1)} + \sin t\, \underset{\sim}{\mathbf{s}}_2^{(1)}\right). \tag{6}$$

Upon choosing the eigenbasis of $\underset{\sim}{\mathbf{s}}_3^{(1)} \otimes \underset{\sim}{\mathbf{s}}_3^{(2)}$ symbolically written as $(\uparrow\uparrow, \uparrow\downarrow, \downarrow\uparrow, \downarrow\downarrow)$ this Hamiltonian can be identified with the Hermitean 4×4-matrix:

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$$H(t)=\begin{pmatrix} \frac{\lambda+4}{4} & 0 & \frac{1}{2}f\mathrm{e}^{-\mathrm{i}t} & 0 \\ 0 & -\frac{\lambda}{4} & \frac{\lambda}{2} & \frac{1}{2}f\mathrm{e}^{-\mathrm{i}t} \\ \frac{1}{2}f\mathrm{e}^{\mathrm{i}t} & \frac{\lambda}{2} & -\frac{\lambda}{4} & 0 \\ 0 & \frac{1}{2}f\mathrm{e}^{\mathrm{i}t} & 0 & \frac{\lambda-4}{4} \end{pmatrix}. \tag{7}$$

First we will solve the corresponding Schrödinger equation ($\hbar=1$)

$$\mathrm{i}\,\frac{\partial}{\partial t}\psi(t)=H(t)\,\psi(t). \tag{8}$$

To this end we differentiate (8) three times w.r.t. t and eliminate all components of $\psi(t)$ except the first one $\psi_1(t)$. This yields a linear 4th order differential equation for $\psi_1(t)$ of the form:

$$\begin{aligned}\frac{\partial^4}{\partial t^4}\psi_1(t)=&-\frac{1}{256}(2f-\lambda-4)(2f+\lambda+4)\left(4f^2+(\lambda+4)(3\lambda-4)\right)\psi_1(t)\\ &-\frac{\mathrm{i}}{8}\left(\left(8f^2+(\lambda-2)(\lambda+4)^2\right)\frac{\partial}{\partial t}\psi_1(t)-\mathrm{i}\left(4f^2+3\lambda^2-48\right)\frac{\partial^2}{\partial t^2}\psi_1(t)+32\frac{\partial^3}{\partial t^3}\psi_1(t)\right).\end{aligned} \tag{9}$$

Remarkably, the coefficients of this differential equations are independent of t due to the circularly polarized form of the driving. In contrast to the present case, for a linearly polarized driving of an $s=1/2$ spin the analogous elimination of the second component of $\psi(t)$ leads to a 2^{nd} order differential equation with t-dependent coefficients. Although this equation can be transformed into a confluent Heun equation, see [31, 32], and [33], it is by far more intricate than the 4th order differential equation obtained in this paper.

In our case the differential equation (9) can be elementarily solved by an exponential ansatz

$$\psi_1(t)=\sum_{n=1}^{4}c_n\,\exp\left(\mathrm{i}\,\omega_n\,t\right), \tag{10}$$

with arbitrary coefficients $c_n\in\mathbb{C}$. The ω_n can be obtained as the roots of an equation of 4th order and assume the form:

$$\omega_1=\frac{1}{4}\left(-2\sqrt{f^2+\lambda^2}+\lambda-4\right), \tag{11}$$

$$\omega_2=\frac{1}{4}\left(2\sqrt{f^2+\lambda^2}+\lambda-4\right), \tag{12}$$

$$\omega_3=\frac{1}{4}(-2f-\lambda-4), \tag{13}$$

$$\omega_4=\frac{1}{4}(2f-\lambda-4). \tag{14}$$

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If we would have included more parameters in the Hamiltonian (6), e.g. the frequency ω of the periodic driving, this result would still be valid, albeit with a more complicated form of the roots that practically rules out a further analytical treatment of the problem.

The remaining three components of $\psi(t)$ are obtained by means of the following equations previously used for eliminating $\psi_2(t), \psi_3(t), \psi_4(t)$:

$$\psi_2(t) = -\frac{\mathrm{e}^{\mathrm{i}\,t}}{4\,f\,\lambda}\left(16\left(2\,\mathrm{i}\,\frac{\partial\psi_1}{\partial t} + \frac{\partial^2\psi_1}{\partial t^2}\right) + \left(-16 + 4f^2 + \lambda^2\right)\psi_1\right), \tag{15}$$

$$\psi_3(t) = \frac{\mathrm{i}\,\mathrm{e}^{\mathrm{i}\,t}}{2\,f}\left(4\,\frac{\partial\psi_1}{\partial t} + \mathrm{i}(\lambda+4)\psi_1\right), \tag{16}$$

$$\psi_4(t) = \frac{\mathrm{e}^{2\,\mathrm{i}\,t}}{8\,f^2\,\lambda}\left(-4\mathrm{i}\left(\frac{\partial\psi_1}{\partial t}\left(4f^2+5\lambda^2+8\lambda-48\right) - 4\mathrm{i}(\lambda-12)\frac{\partial^2\psi_1}{\partial t^2} + 16\frac{\partial^3\psi_1}{\partial t^3}\right.\right.$$
$$\left.\left. + \left((\lambda+4)^2(3\lambda-4) - 4f^2(\lambda-4)\right)\psi_1\right)\right). \tag{17}$$

Inserting $\psi_1(t)$ according to (10) and (11)–(14) into (15)–(17) yields a first solution $\psi^{(1)}(t)$ that will be rewritten as

$$\psi^{(1)}(t) = U(t)\begin{pmatrix} c_1 \\ c_2 \\ c_3 \\ c_4 \end{pmatrix}, \tag{18}$$

where $U(t)$ is a unitary 4×4-matrix satisfying

$$\frac{\partial}{\partial t}U(t) = -\mathrm{i}\,H(t)\,U(t). \tag{19}$$

From this we obtain the fundamental system of solutions $\Psi(t)$ by

$$\Psi(t) \equiv U(t)\,U(0)^{-1}, \tag{20}$$

satisfying

$$\Psi(0) = \mathbb{1}. \tag{21}$$

We will only explicitly give $\Psi(t)$ in its *Floquet normal form*

$$\Psi(t) = \mathcal{P}(t)\,\mathrm{e}^{-\mathrm{i}\mathcal{F}\,t}, \tag{22}$$

such that $\mathcal{P}(t)$ is 2π-periodic and $\mathcal{F}$ is the Floquet matrix. After some calculations we obtain

$$\mathcal{P}(t) = \begin{pmatrix} \mathrm{e}^{-\mathrm{i}\,t} & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & \mathrm{e}^{\mathrm{i}\,t} \end{pmatrix}, \tag{23}$$

and

$$e^{-i\mathcal{F}t} = A\,\Delta(t)\,A^{\top}, \tag{24}$$

where

$$A = \frac{1}{2}\begin{pmatrix} -\sqrt{1-\alpha} & 1 & \sqrt{1+\alpha} & 1 \\ -\sqrt{1+\alpha} & -1 & -\sqrt{1-\alpha} & 1 \\ \sqrt{1+\alpha} & -1 & \sqrt{1-\alpha} & 1 \\ \sqrt{1-\alpha} & 1 & -\sqrt{1+\alpha} & 1 \end{pmatrix}, \tag{25}$$

setting

$$\alpha \equiv \frac{\lambda}{\sqrt{f^2+\lambda^2}}, \tag{26}$$

and

$$\Delta(t) = \begin{pmatrix} e^{\frac{1}{4}it\left(2\sqrt{f^2+\lambda^2}+\lambda\right)} & 0 & 0 & 0 \\ 0 & e^{\frac{1}{4}it\,(2f-\lambda)} & 0 & 0 \\ 0 & 0 & e^{\frac{1}{4}it\left(\lambda-2\sqrt{f^2+\lambda^2}\right)} & 0 \\ 0 & 0 & 0 & e^{-\frac{1}{4}it\,(2f+\lambda)} \end{pmatrix}. \tag{27}$$

The connection to the Floquet functions $u_n(t)$ mentioned in the Introduction is given by

$$u_n(t) = \mathcal{P}(t)\,A_n, \tag{28}$$

where A_n denotes the nth column of A.

We note the following special features of the form of (22) not yet fully understood. First, it is not *a priori* clear that according to (23) the periodic part $\mathcal{P}(t)$ is diagonal in the spin basis and hence $[\mathcal{P}(t_1), \mathcal{P}(t_2)] = 0$ for all $t_1,\, t_2 \in \mathbb{R}$. Second, the eigenvectors of the Floquet matrix $\mathcal{F}$ that are the columns of A according to (25) are real. This follows also from the fact the monodromy matrix $\Psi(2\pi)$ is unitary and symmetric, the latter property being a consequence of the particular structure of the Hamiltonian (7), see appendix A. Note also that the second and the fourth eigenvector is independent of f and λ. These special properties of the monodromy matrix may explain the occurrence of the phase boundaries described in section 4.2 despite the effect of 'avoided level crossing', see also the corresponding remarks in appendix A.

The quasienergies ϵ_n (eigenvalues of $\mathcal{F}$) can be directly read off the diagonal elements of (27) that represent the eigenvalues of $e^{-i\mathcal{F}t}$:

$$\epsilon_1 = -\frac{1}{4}\left(2\sqrt{f^2+\lambda^2}+\lambda\right), \tag{29}$$

$$\epsilon_2 = \frac{1}{4}(\lambda-2f). \tag{30}$$

$$\epsilon_3 = \frac{1}{4}\left(2\sqrt{f^2+\lambda^2}-\lambda\right) \tag{31}$$

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$$\epsilon_4 = \frac{1}{4}(\lambda + 2f). \tag{32}$$

Recall that the quasienergies are uniquely determined only up to integer multiples of $\omega = 1$. In (29)–(32) we have chosen representatives of quasienergies that appear in a strictly monotonic increasing order for $\lambda,\ f > 0$ which facilitates the calculations in the periodic thermodynamics section 4.2. For the sake of consistency we will check the two limits $\lambda \to 0$ and $f \to 0$.

The static limit $f \to 0$ yields

$$\lim_{f\to 0} \epsilon_2 = \lim_{f\to 0} \epsilon_3 = \lim_{f\to 0} \epsilon_4 = \frac{\lambda}{4}, \quad \text{and} \quad \lim_{f\to 0} \epsilon_1 = -\frac{3\lambda}{4}. \tag{33}$$

This agrees with the eigenvalues (5) of the static Hamiltonian H_0 modulo integers.

The limit $\lambda \to 0$ means that the two spins are decoupled and hence the quasienergies should approach those of the usual Rabi problem for the first spin plus the energy eigenvalues $\pm\frac{1}{2}$ of the second spin. We obtain

$$\lim_{\lambda\to 0} \epsilon_3 = \lim_{\lambda\to 0} \epsilon_4 = \frac{f}{2}, \quad \text{and} \quad \lim_{\lambda\to 0} \epsilon_1 = \lim_{\lambda\to 0} \epsilon_2 = -\frac{f}{2}. \tag{34}$$

This has to be compatible with

$$\epsilon_{\text{Rabi}} = \frac{\omega \pm \Omega}{2}, \tag{35}$$

where Ω is the Rabi frequency

$$\Omega = \sqrt{f^2 + (\omega_0 - \omega)^2}. \tag{36}$$

In our case we have chosen $\omega_0 = \omega = 1$ which implies $\Omega = f$ and further $\epsilon_{\text{Rabi}} = \frac{1 \pm f}{2}$. The total quasienergy of the decoupled spin system is thus $\epsilon = \frac{1\pm f}{2} \pm \frac{1}{2}$. Again, this is, modulo integers, in accordance with (34).

3. Work performed on a two spin system

As an application of the results obtained in the preceding section 2 we consider the work performed on a two level system by a circularly polarized magnetic field during one period. In contrast to classical physics this work is not just a number but, following [34], has to be understood in terms of two subsequent energy measurements. Before the time $t = 0$ the two level system is assumed to be in a mixed state according to the canonical ensemble

$$W = \exp\left(-\beta H_0\right) / \text{Tr}\left(\exp\left(-\beta H_0\right)\right), \tag{37}$$

with dimensionless inverse temperature $\beta = \frac{\hbar\omega}{k_B T}$ and H_0 being the static Hamiltonian (4). Then at the time $t = 0$ one performs a Lüders measurement of the instantaneous energy H_0 with the four possible outcomes E_n, $n = 1, \ldots, 4$ according to (5). Hence after the measurement the system is in the pure state P_n with probability

$\mathrm{Tr}\,(P_n W) = \frac{1}{Z}\mathrm{e}^{-\beta E_n}$, $n = 1, \ldots, 4$, where the P_n are the projectors onto the eigenstates of H_0, i.e.

$$P_1 = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}, \quad P_2 = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \quad P_3 = \frac{1}{2}\begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 1 & -1 & 0 \\ 0 & -1 & 1 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}, \quad P_4 = \frac{1}{2}\begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 1 & 1 & 0 \\ 0 & 1 & 1 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}, \tag{38}$$

and $Z = \sum_{n=1}^{4} \mathrm{e}^{-\beta\, E_n}$. After this measurement the system evolves according to the Schrödinger equation (8) with Hamiltonian $H(t)$. At the time $t = 2\pi$ the system hence is in the pure state $\Psi(2\pi)\, P_n\, \Psi(2\pi)^*$ with probability $\mathrm{Tr}\,(P_n W)$ for $n = 1, \ldots, 4$. Then a second measurement of the static energy H_0 is performed, again with the four possible outcomes E_n. Both measurements together have $4 \times 4 = 16$ possible outcomes symbolized by pairs (i, j) where $i, j = 1, \ldots 4$ that occur with probabilities

$$p(i,j) = \mathrm{Tr}\,(W\, P_i)\, \mathrm{Tr}\,(P_j\, \Psi(2\pi)\, P_i\, \Psi(2\pi)^*)\,, \tag{39}$$

such that $\sum_{i,j=1}^{4} p(i,j) = 1$. We will not display the $p(i,j)$ but rather the marginal probabilities $p(i) \equiv \sum_{j=1}^{4} p(i,j)$ and the conditional probabilities $\pi(\,j|i) \equiv \frac{p(i,j)}{p(i)}$, the latter being independent of β. It is plausible and can be directly verified that the matrix of conditional probabilities will be symmetric and hence doubly stochastic, see [35] for the rôle of double stochasticity in connection with the Jarzynski equation. Thus we need only to display the values of $\pi(\,j|i)$ for $j \leqslant i$. The detailed results are

$$p(1) = \frac{1}{z}\mathrm{e}^{2\beta}, \quad p(2) = \frac{1}{z}\mathrm{e}^{\beta(1+\lambda)}, \quad p(3) = \frac{1}{z}\mathrm{e}^{\beta}, \quad p(4) = \frac{1}{z} \equiv \frac{1}{\mathrm{e}^{\beta}\,(\mathrm{e}^{\beta\lambda} + \mathrm{e}^{\beta} + 1) + 1}, \tag{40}$$

and

$$\pi(1|1) = \pi(4|4) = a + b, \tag{41}$$

$$\pi(1|4) = a - b, \tag{42}$$

$$a = \frac{1}{8}\left(\frac{f^2\cos\left(2\pi\sqrt{f^2+\lambda^2}\right) + 2f^2 + 3\lambda^2}{f^2+\lambda^2} + \cos(2\pi f)\right), \tag{43}$$

$$b = \frac{1}{2}\cos(\pi f)\left(\frac{\lambda\sin(\pi\lambda)\sin\left(\pi\sqrt{f^2+\lambda^2}\right)}{\sqrt{f^2+\lambda^2}} + \cos(\pi\lambda)\cos\left(\pi\sqrt{f^2+\lambda^2}\right)\right), \tag{44}$$

$$\pi(1|2) = \pi(2|4) = \frac{f^2\sin^2\left(\pi\sqrt{f^2+\lambda^2}\right)}{2\,(\,f^2+\lambda^2)}, \tag{45}$$

$$\pi(2|2) = \frac{f^2\cos\left(2\pi\sqrt{f^2+\lambda^2}\right) + f^2 + 2\lambda^2}{2\,(\,f^2+\lambda^2)}, \tag{46}$$

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$$\pi(2|3) = 0. \tag{47}$$

Besides the symmetry of the matrix of conditional probabilities there are additional coincidences in (41), (45) and vanishing values in (47) that are not yet understood.

The matrix of probabilities $p(i,j)$ contains all information for the probability distribution of the energy differences between the first and the second measurement, i.e. of the distribution of the work w performed on the two spin system by means of the periodic driving. Interestingly, although 'work' cannot be considered as an observable in the ordinary sense giving rise to a projection-valued measure [34], it is an observable in the generalized sense of a positive-operator-valued measure [36, 37].

For example, we may calculate the mean value of the performed work with the result

$$\langle w \rangle = \sum_{i,j=1}^{4} (E_j - E_i)\, p(i,j) = \frac{1}{4(f^2+\lambda^2)z}\, (w_1 + w_2 + w_3)\,, \tag{48}$$

$$w_1 = 4\left(\mathrm{e}^{2\beta} - 1\right)\lambda^2 - f^2\left(\mathrm{e}^{2\beta}(\lambda - 4) - 2\lambda \mathrm{e}^{\beta\lambda+\beta} + \lambda + 4\right), \tag{49}$$

$$\begin{aligned} w_2 = {} & f^2\lambda\left(-2\mathrm{e}^{\beta\lambda+\beta} + \mathrm{e}^{2\beta} + 1\right)\cos\left(2\pi\sqrt{f^2+\lambda^2}\right) \\ & - 8\mathrm{e}^{\beta}\sinh(\beta)\left(f^2+\lambda^2\right)\cos(\pi f)\cos(\pi\lambda)\cos\left(\pi\sqrt{f^2+\lambda^2}\right) \end{aligned} \tag{50}$$

$$w_3 = -4\left(\mathrm{e}^{2\beta} - 1\right)\lambda\sqrt{f^2+\lambda^2}\cos(\pi f)\sin(\pi\lambda)\sin\left(\pi\sqrt{f^2+\lambda^2}\right), \tag{51}$$

where the parameter z in (48) has been defined in (40). This function is shown in figure 2 for the inverse temperature $\beta = 1$. First, we note that obviously $\langle w \rangle \geqslant 0$ which appears physically plausible and will be proven below.

Another conspicuous feature of the graph of $\langle w \rangle(\lambda, f, 1)$ is its oscillating behaviour with increasing amplitude for large values of $\lambda \approx f$. This will be more clearly demonstrated in the figure 3 where we have set $\lambda = f$ and displayed $\langle w \rangle(f, f, \beta)$ for values of $\beta = 0, 1, \ldots, 20$. It is obvious from this figure and can be analytically confirmed that

$$\langle w \rangle(f, f, \beta) \sim \frac{1}{2} f \sin^2\left(\sqrt{2}\,\pi\, f\right) \text{ for } f \to \infty. \tag{52}$$

The convergence of $\langle w \rangle(f, f, \beta)$ against its asymptotic behaviour holds for all $\beta \geqslant 0$ but will be more rapid for large β. We will give a semi-quantitative explanation. For large β, i.e. low temperatures the system is practically in its ground state with energy E_3 at $t = 0$, the begin of the periodic driving, see (5). By the driving it will be excited to the next lowest state with energy E_2. The probability of excitation $p_{3\to 2}(t)$ can be calculated and yields a rather simple expression for the special case $\lambda = f$:

$$p_{3\to 2}(t) = \frac{1}{4}\sin^2\left(\frac{f\,t}{\sqrt{2}}\right). \tag{53}$$

This result is analogous to the well-known Rabi oscillation of a two-level system. It is further plausible that the mean value of the work during one period will be maximal

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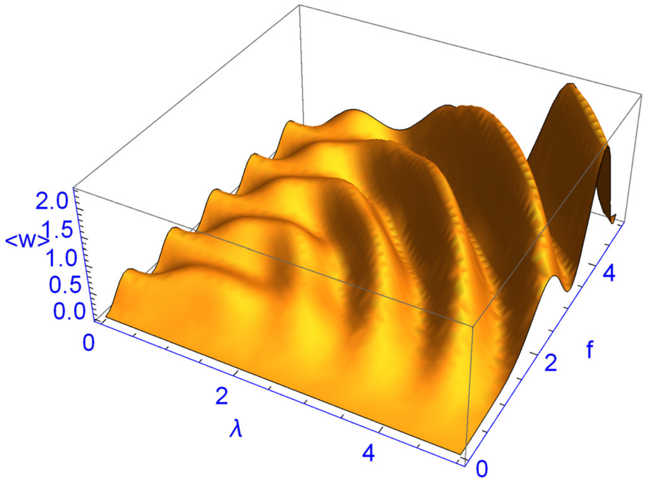

Figure 2. The mean value $\langle w \rangle$ of the work performed on the two spin Rabi system during one period as a function of the physical parameters λ and f, where the initial inverse temperature of the system has been set to $\beta = 1$.

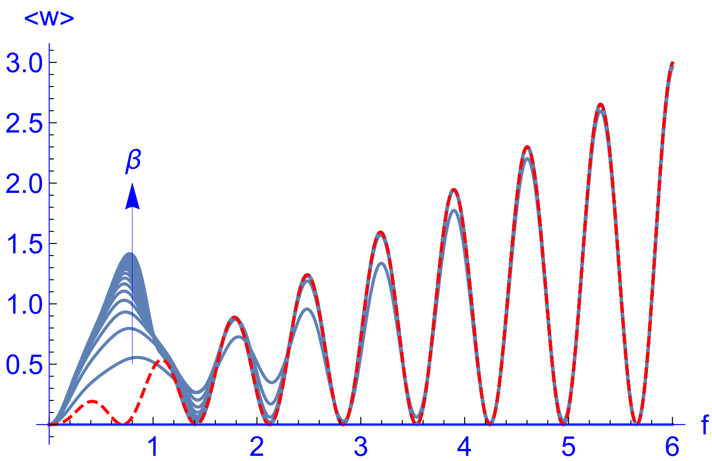

Figure 3. The mean value $\langle w \rangle$ of the work performed on the two spin Rabi system during one period as a function of the physical parameters $\lambda = f$ and $\beta = 0, 1, \ldots, 20$, where the increasing values of β are indicated by an arrow. Moreover, we show the asymptotic form of $\langle w \rangle \sim \frac{1}{2} f \sin^2\left(\sqrt{2}\,\pi\, f\right)$ (red, dashed curve).

if some maximum of (53) will be attained after exactly one period of driving, i.e. at $t = 2\pi$. This happens for

$$\frac{f\,2\,\pi}{\sqrt{2}} = \frac{n\,\pi}{2}, \quad n \text{ being odd} \quad \Leftrightarrow f = \frac{n}{2\sqrt{2}}, \tag{54}$$

and hence at the maxima of the asymptotic form of $\langle w \rangle(f, f, \beta) \sim \frac{1}{2} f \sin^2\left(\sqrt{2}\,\pi\, f\right)$. An analogous reasoning applies to the minima of $\langle w \rangle(f, f, \beta)$ Hence the oscillating structure of $\langle w \rangle$ visible in the figure 2 can be viewed as a footprint of a kind of approximate Rabi oscillation occurring for the two spin Rabi model. Moreover, it is also plausible that asymptotically $\langle w \rangle(f, f, \beta)$ scales with f.

Finally, we may, after some calculations, confirm the famous Jarzynski equation [34] that in our case reads

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$$\left\langle \mathrm{e}^{-\beta\, w} \right\rangle = \sum_{i,j=1}^{4} \mathrm{e}^{-\beta\,(E_j - E_i)}\, p(i,j) = 1. \tag{55}$$

The latter can be considered as a test of consistency of our results. Further, we may apply Jensen's inequality to the convex function $x \mapsto -\log x$ and conclude

$$\langle \beta\, w \rangle = \left\langle -\log\left(\mathrm{e}^{-\beta\, w}\right)\right\rangle \overset{\text{Jensen}}{\geqslant} -\log\left\langle \mathrm{e}^{-\beta\, w}\right\rangle \overset{(55)}{=} -\log 1 = 0, \tag{56}$$

which, due to $\beta > 0$, means that the expectation value of the performed work is always non-negative which would be difficult to be confirmed directly for the expression (48)–(51) of $\langle w \rangle$.

4. Periodic thermodynamics

4.1. Golden-rule approach to open driven systems

Let us consider a quantum system evolving according to a $T = \frac{2\pi}{\omega}$-periodic Hamiltonian $H(t)$ on a Hilbert space $\mathcal{H}_S$ that is additionally coupled to a heat bath, described by a Hamiltonian H_{bath} acting on a Hilbert space $\mathcal{H}_B$. The total Hamiltonian on the composite Hilbert space $\mathcal{H}_S \otimes \mathcal{H}_B$ takes the form

$$H_{\text{total}}(t) = H(t) \otimes \mathbb{1} + \mathbb{1} \otimes H_{\text{bath}} + V \otimes W\ . \tag{57}$$

Moreover, following Breuer *et al* [11], let us consider a bath consisting of thermally occupied harmonic oscillators, and an interaction of the prototypical form

$$W = \sum_{\widetilde{\omega}} \left(b_{\widetilde{\omega}} + b_{\widetilde{\omega}}^{\dagger} \right)\ , \tag{58}$$

where $b_{\widetilde{\omega}}$ ($b_{\widetilde{\omega}}^{\dagger}$) is the annihilation (creation) operator pertaining to a bath oscillator of frequency $\widetilde{\omega}$.

For weak coupling the effect of the heat bath can be approximately described by a variant of the Golden Rule. Since this approach has been elaborately explained in the literature, see [25] and [29], we will confined ourselves here with the enumeration of the pertinent formulas sticking closely to [29].

In the golden-rule approximation the heat bath induces transitions between the system's Floquet states $u_i(t)$ and $u_f(t)$ with transition rates Γ_{fi} that can be written as sums over partial rates

$$\Gamma_{fi} = \sum_{\ell \in \mathbb{Z}} \Gamma_{fi}^{(\ell)} \tag{59}$$

given by

$$\Gamma_{fi}^{(\ell)} = 2\pi\, |V_{fi}^{(\ell)}|^2\, N(\omega_{fi}^{(\ell)})\, J(|\omega_{fi}^{(\ell)}|)\ . \tag{60}$$

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Here $J(|\omega_{fi}^{(\ell)}|)$ denotes the spectral density of the frequency of bath phonons and will be set to a constant $J_0 > 0$ in what follows. Further, $V_{fi}^{(\ell)}$ denotes the Fourier components of the T-periodic matrix elements

$$\tilde{V}_{fi} = \langle u_f(t)| V |u_i(t)\rangle = \sum_{\ell\in\mathbb{Z}} V_{fi}^{(\ell)} \exp(\mathrm{i}\ell\omega t) \ , \tag{61}$$

and $N(\omega_{fi}^{(\ell)})$ is the value of the function $N(\tilde{\omega})$ evaluated at

$$\omega_{fi}^{(\ell)} \equiv \epsilon_f - \epsilon_i + \ell\,\omega. \tag{62}$$

Physically, $N(\tilde{\omega})$ represents the thermal average of the bath phonon occupation density and is given by

$$N(\tilde{\omega}) = \begin{cases} \tilde{\omega} > 0 & : \quad \frac{1}{\exp(\beta\tilde{\omega})-1}, \\ \tilde{\omega} < 0 & : \quad \frac{1}{1-\exp(\beta\tilde{\omega})}, \end{cases} \tag{63}$$

where β is the inverse temperature of the bath, not to be confounded with the inverse temperature considered in section 3. The case distinction in (63) corresponds to the distinction between the creation of a bath phonon ($\tilde{\omega} > 0$) and its absorption ($\tilde{\omega} < 0$). Thus, a transition among Floquet states is not simply associated with only one single frequency, but rather with a set of frequencies spaced by integer multiples of the driving frequency ω, reflecting the ladder-like nature of the system's quasienergies.

The total rates (59) now determine the desired quasi-stationary distribution $\{p_n\}$ as a solution to the Pauli master equation [11]

$$\sum_m \left(\Gamma_{nm} p_m - \Gamma_{mn} p_n\right) = 0 \ , \tag{64}$$

where the existence of a strictly positive solution will be shown below. According to this equation (64), the quasi-stationary distribution $\{p_m\}$ which establishes itself under the combined influence of time-periodic driving and the thermal oscillator bath is the eigenvector of a matrix $\widetilde{\Gamma}$ corresponding to the eigenvalue 0, where $\widetilde{\Gamma}$ is obtained from Γ by subtracting from the diagonal elements the respective column sums, i.e.

$$\widetilde{\Gamma}_{mn} \equiv \Gamma_{mn} - \delta_{mn} \sum_{k=1}^{N} \Gamma_{kn} \ . \tag{65}$$

Moreover, it is evident that we only need the non-diagonal matrix elements of Γ for calculating the quasistationary distribution, whereas the diagonal elements would be required for computing the dissipation rate [25].

As announced above, we will now prove the existence of a strictly positive solution of the Pauli master equation (64). Although this result it well-known it is not easily found in the literature and hence an explicit proof will be in order.

We start with a few definitions needed for the statement of the theorem of Frobenius–Perron that is suited for the problem at hand. A real $N \times N$-matrix T will be called *non-negative*, in symbols $T \geqslant 0$, iff all its matrix entries satisfy $T_{ij} \geqslant 0$. Analogously, we will define a *positive* matrix $T > 0$ and also use these terms for vectors x with the notation $x > 0$ or $x \geqslant 0$. Moreover, T is *irreducible* iff for all $1 \leqslant i, j \leqslant N$ there exists a

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$k \in \mathbb{N}$ such that $T_{ij}^k > 0$. Physically, if T is some transition matrix, the notion of irreducibility would be construed as a kind of 'ergodicity', because it says that if starting from any state i it is possible to reach any other state j after a finite number of steps. Then we may state the theorem of Frobenius–Perron, see, e.g. [38], Theorem 2, p. 53, in the following form, adapted to our purposes.

Theorem 1 (Frobenius–Perron). *Let T be a non-negative irreducible square matrix. Then*

- *T has a positive eigenvalue $\lambda_{\max}$ that is the spectral radius of T, i.e. all other eigenvalues λ of T satisfy $|\lambda| \leqslant \lambda_{\max}$.*
- *Furthermore $\lambda_{\max}$ has algebraic and geometric multiplicity one, and has an eigenvector x with $x > 0$.*
- *Any non-negative eigenvector of T is a multiple of x.*

By means of (60) it is obvious that $\Gamma \geqslant 0$, but the present two spin Rabi model is an example showing that $\Gamma > 0$ does not hold in general, see below. Hence, in order to apply the preceding theorem, we will additionally need the following

Assumption 1. *Γ is irreducible.*

That is essentially saying that the eigenvectors of the interaction matrix V are oblique w.r.t. the Floquet basis and does not follow from the general assumptions made so far.

Recall that $\widetilde{\Gamma}$ is defined by subtraction of the column sums of Γ and hence will possess negative matrix entries in the diagonal. If λ is defined as the maximal column sum of Γ we will obtain a non-negative matrix G by adding λ to each diagonal element,

$$G \equiv \widetilde{\Gamma} + \lambda \mathbb{1} \geqslant 0, \tag{66}$$

and, moreover, conclude

Lemma 1. *G and hence also $G^\top$ are irreducible.*

Proof. By definition, G can be written as $G = \Gamma + \Delta$ such that $\Delta \geqslant 0$ is a diagonal matrix. It follows from

$$G^k = (\Gamma + \Delta)^k = \Gamma^k + \Delta\,\Gamma^{k-1} + \ldots + \Gamma^{k-1}\Delta + \ldots + \Delta^2\Gamma^{k-2} + \ldots + \Gamma^{k-2}\Delta^2 + \ldots + \Delta^k, \tag{67}$$

and the assumption 1 that for all $1 \leqslant i, j \leqslant N$ there exists a $k \in \mathbb{N}$ such that $G_{ij}^k > 0$. Hence G is irreducible. $\square$

By definition, $\widetilde{\Gamma}$ has vanishing column sums, hence $\mathbf{1} \equiv (1, 1, \ldots, 1)$ will be a left eigenvector of $\widetilde{\Gamma}$ with eigenvalue 0. It follows that $\mathbf{1}$ is also a right eigenvector of $G^\top$ with eigenvalue λ. $G^\top$ satisfies the conditions of the theorem of Frobenius–Perron, hence $\lambda = \lambda_{\max}$ is the spectral radius of $G^\top$ and $\mathbf{1}$ is the unique corresponding eigenvector. Applying again the theorem of Frobenius–Perron to G that has the same eigenvalues as $G^\top$ we conclude that there exists an eigenvector $p > 0$ of G with eigenvalue λ, unique up to normalization. It follows that $\widetilde{\Gamma}\, p = 0$ and hence p is the solution of the Pauli master equation (64) we are seeking for. We state this result as

Theorem 2. *If the matrix Γ is irreducible then the Pauli master equation (64) has a unique solution $\{p_n\}$ satisfying $p_n > 0$ for all $n = 1, \ldots N$ and $\sum_{n=1}^{N} p_n = 1$.*

4.2. Application to the two spin system

We choose the matrix V that is part of the coupling to the heat bath according to (57) as $V \equiv \mathbb{1} \otimes \mathbf{s}_1^{(2)}$, i.e. only the second spin is involved. We need its matrix elements $\tilde{V}_{fi} \equiv \langle u_f(t)|\, V\, |u_i(t)\rangle$ w.r.t. Floquet states, see (61). In our case $\tilde{V}$ can be written as

$$\tilde{V} = A^*\, \mathcal{P}(t)^*\, V\, \mathcal{P}(t)\, A, \tag{68}$$

with $\mathcal{P}(t)$ and A according to (23) and (25). It is clear from (23) that $\tilde{V}$ contains only Fourier components of the order $|\ell| \leqslant 1$. Actually, we obtain

$$\tilde{V} = V^{(1)}\, \mathrm{e}^{\mathrm{i}t} + V^{(-1)}\, \mathrm{e}^{-\mathrm{i}t}, \tag{69}$$

where

$$V^{(1)} = \frac{1}{8} \begin{pmatrix} 2fu & v+w & -2\lambda u & v-w \\ -v-w & -2 & v-w & 0 \\ -2\lambda u & w-v & -2fu & v+w \\ w-v & 0 & -v-w & 2 \end{pmatrix}, \tag{70}$$

$$V^{(-1)} = \frac{1}{8} \begin{pmatrix} 2fu & -v-w & -2\lambda u & w-v \\ v+w & -2 & w-v & 0 \\ -2\lambda u & v-w & -2fu & -v-w \\ v-w & 0 & v+w & 2 \end{pmatrix}, \tag{71}$$

and

$$u \equiv \frac{1}{\sqrt{f^2+\lambda^2}}, \quad v \equiv \sqrt{1+\lambda u}, \quad w \equiv \sqrt{1-\lambda u}. \tag{72}$$

Note that the occurrence of the matrix entry 0 in (70) and (71) implies that $\Gamma_{24} = \Gamma_{42} = 0$ and hence Γ is not positive but only non-negative which has to be taken into account in the application of theorem 1.

Further we need the values of $N(\omega_{fi}^{(\ell)})$ in (60) according to (63). Recall that the case distinction to be made w.r.t. the sign of $\omega_{fi}^{(\ell)} = \epsilon_f - \epsilon_i + \ell\omega = \epsilon_f - \epsilon_i + \ell$ physically corresponds to the absorption or generation of bath phonons. In order to obtain analytical expressions for, say, the occupation probabilities in the non-equilibrium steady state (NESS), we will have to restrict the parameters $(\lambda, f) \in \mathbb{R}_+ \times \mathbb{R}_+$ to certain domains where the sign of $\omega_{fi}^{(\ell)}$ will not change for all f, i, ℓ. These domains can be viewed as 'phases' of a phase diagram of the parameter space $\mathbb{R}_+ \times \mathbb{R}_+$. The boundaries of these phases are given by equations of the form $\omega_{fi}^{(\ell)} = 0$. The latter corresponds to a partial

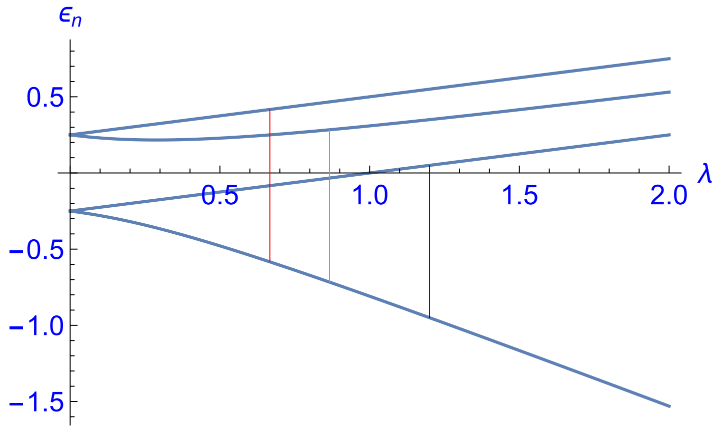

Figure 4. The four quasienergies ϵ_n according to (29)–(32) as functions of λ where f has been set to $1/2$. At the values of $\lambda = \frac{2}{3}, \frac{\sqrt{3}}{2}$ and $\frac{6}{5}$ certain differences of quasienergies assume the value 1 and hence the corresponding frequencies $\omega_{fi}^{(\ell)}$ according to (62) vanish. These cases are indicated by vertical coloured lines. They correspond to certain phase boundaries in figure 6.

degeneracy of quasienergies taking into account that they are only defined up to integer multiples of the driving frequency $\omega = 1$.

We consider the example $f = 3$, $i = 1$, and $\ell = -1$. The corresponding boundary equation is

$$0 = \omega_{31}^{(-1)} = \epsilon_3 - \epsilon_1 - 1 = \frac{1}{4}\left(-\lambda + 2\sqrt{f^2 + \lambda^2}\right) + \frac{1}{4}\left(\lambda + 2\sqrt{f^2 + \lambda^2}\right) - 1 = \sqrt{f^2 + \lambda^2} - 1, \tag{73}$$

describing a quarter circle in the (λ, f)-quadrant, see figure 6.

The other boundaries are given by

$$0 = \epsilon_2 - \epsilon_1 - 1 \Leftrightarrow f = \frac{2(\lambda - 1)}{2 - \lambda}, \tag{74}$$

$$0 = \epsilon_3 - \epsilon_2 - 1 \Leftrightarrow f = \frac{2(\lambda + 1)}{2 + \lambda}, \tag{75}$$

$$0 = \epsilon_4 - \epsilon_1 - 1 \Leftrightarrow f = \frac{2(\lambda - 1)}{\lambda - 2}, \tag{76}$$

$$0 = \epsilon_4 - \epsilon_2 - 1 \Leftrightarrow f = 1 \tag{77}$$

see the figures 4–6. Note that there are six positive differences of quasienergies $\epsilon_f - \epsilon_i$ but only five boundary equations since the equation $\epsilon_4 - \epsilon_3 - 1 = 0$ has no positive solution.

As a first, somewhat surprising analytical result we note that for the phase A defined by $f < \frac{2(\lambda-1)}{\lambda-2}$, see figure 6, the Pauli master equation (64) has a unique solution corresponding to the same occupation probability for all Floquet states. This also follows from the symmetry $\Gamma_{mn} = \Gamma_{nm}$ that holds only within phase A. Formally the coincidence of all probabilities would correspond to an infinite quasitemperature and

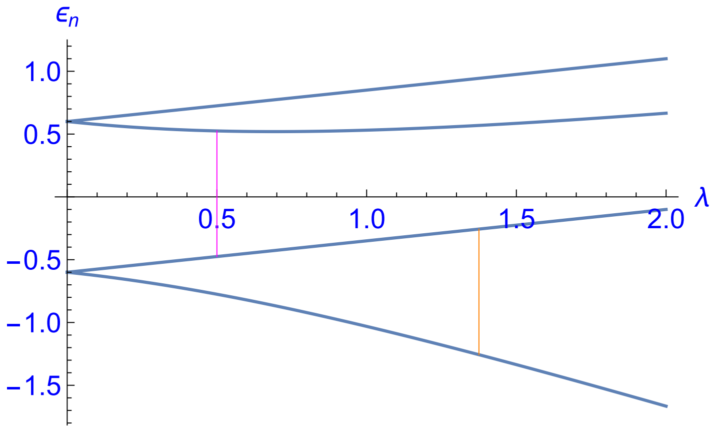

Figure 5. Analogous to figure 4 but with $f = \frac{6}{5}$. Here the frequencies $\omega_{fi}^{(\ell)}$ vanish at $\lambda = \frac{1}{2}$ and $\lambda = \frac{11}{8}$.

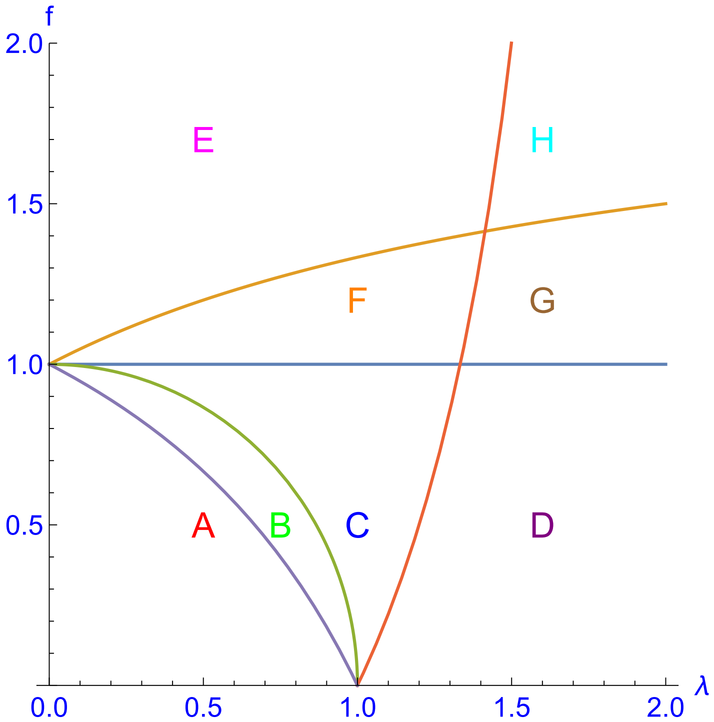

Figure 6. The phase diagram of the (λ, f)-parameter space with eight phases $A, \dots H$, where the phase boundaries are given by the equations (73) (green circle) or (74)–(77) (red, yellow, purple, blue curves).

could be compared with the vanishing inverse quasitemperature along the line $\omega = \omega_0$ and $0 < F < \omega_0$ for the circularly polarized Rabi problem, see [29], figure 1.

In the phase domains B–H the occupation probabilities p_n can be analytically calculated by the means of computer-algebraic software but the results cannot be displayed due to their forbidding complexity. Nevertheless, one may plot these results. A first graphics shows the p_n as continuous functions of λ where the parameter f has been set to $f = 1/2$, see figure 7. One clearly distinguishes the four phases A–D according to figure 6 and observes that the $p_n(\lambda)$ are smooth inside the phase domains but shows

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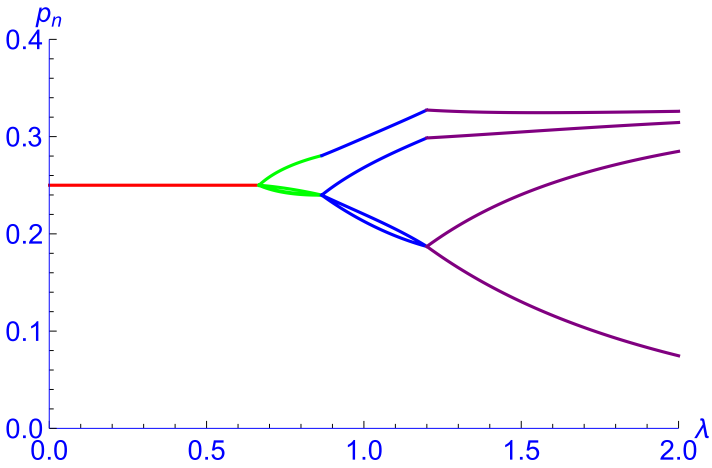

Figure 7. The four occupation probabilities p_n of the Floquet states for the NESS as functions of λ where f has been set to $f = 1/2$ and the inverse bath temperature is chosen as $\beta = 1$. Within the phases A–D, indicated by different colours, the p_n are smooth functions of λ. At the phase boundaries the derivatives $\frac{\mathrm{d}\,p_n}{\mathrm{d}\,\lambda}$ are discontinuous and at least two probabilities coincide.

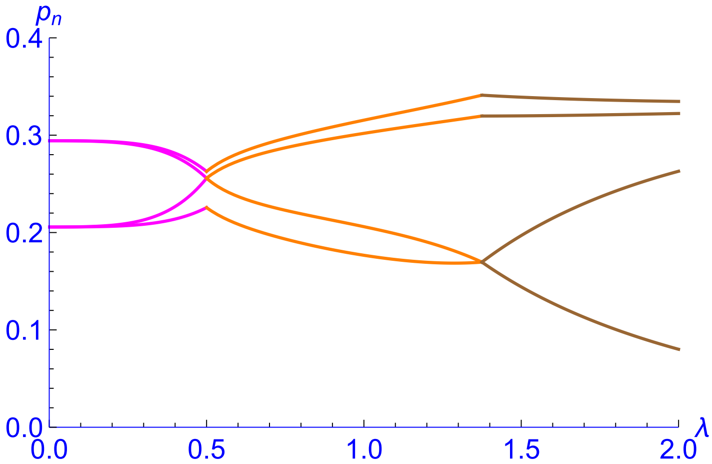

Figure 8. The four occupation probabilities p_n of the Floquet states for the NESS as functions of λ where f has been set to $f = 6/5$ and $\beta = 1$. Within the phases E, F, G, indicated by different colours, the p_n are smooth functions of λ. At the phase boundaries the derivatives $\frac{\mathrm{d}\,p_n}{\mathrm{d}\,\lambda}$ are discontinuous and exactly two probabilities coincide.

kinks at the phase boundaries. The fact that at least two probabilities coincide at the phase boundaries can be understood by the arguments presented in appendix B that also hold for general N-level systems.

The coincidence of two probabilities at phase boundaries also shows that, in general, the NESS will not be of Boltzmann type with a quasitemperature θ: for a Boltzmann distribution of occupation probabilities p_n and non-degenerate representatives of quasienergies two probabilities never coincide except for $\theta = \infty$. In our case the latter only occurs in the phase A, see above.

5. Summary and outlook

We have investigated the two spin Rabi model consisting of an $s = 1/2$ spin subjected to a monochromatic circularly polarized magnetic field and coupled to a second spin $s = 1/2$ that is in turn in contact with a heat bath. The quasienergies of the spin system as well as the occupation probabilities of the emerging NESS can be, in principle, analytically determined and hence this system may serve as an example for testing conjectures about general periodically driven N-level systems. We found that, in contrast to other systems recently studied, the NESS probabilities are not of Boltzmann type and hence there does not exist a quasitemperature. Moreover, the parameter space of the system is found to be partitioned into certain phases such that the NESS probabilities change at the phase boundaries in a way analogous to a 2^{nd} order phase transition. It has been made plausible by detailed arguments that these two properties will also be satisfied for general N-level systems. On the other hand, the existence of a phase A with infinite quasitemperature hinges on special properties of the two spin Rabi model, e.g. the structure of the eigenvectors of the Floquet operator or the commuting operators describing the periodic part of the time evolution, and probably does not generally hold. Nevertheless, it would be instructive to closer investigate similar systems in order to verify (or falsify) the above conjectures.

Acknowledgments

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Appendix A. Proof of the symmetry of the monodromy matrix

As noted in section 2 the symmetry of the unitary monodromy matrix $U(2\pi)$ has the consequence that it possesses a real eigenbasis. In fact, the eigenvalue equation

$$U(2\pi)\,\phi = c\,\phi, \tag{A.1}$$

satisfying $|c|^2 = 1$ implies

$$\overline{\phi} = U(2\pi)\,U(2\pi)^{-1}\,\overline{\phi} = U(2\pi)\,\overline{U(2\pi)}\,\overline{\phi} \overset{(A.1)}{=} \bar{c}\,U(2\pi)\,\overline{\phi}, \tag{A.2}$$

where we have used that, according to the above symmetry assumption, $\overline{U(2\pi)} = U(2\pi)^{-1}$. This means that the vector $\overline{\phi}$ will be an eigenvector of $U(2\pi)$ corresponding to the same eigenvalue $\frac{1}{\bar{c}} = c$. Thus if ϕ is unique it must be real, or otherwise, in the case of degeneracy, it can be chosen as real.

It remains to show that $U(2\pi)$ is symmetric. To this end we introduce a slightly more general notation by writing the unitary time evolution between $t = t_0$ and $t = t_1$ as $U(t_1, t_0)$ such that

$$U(t_1, t_0) = U(t_0, t_1)^{-1}. \tag{A.3}$$

$U(t,0)$ satisfies the differential equation

$$\frac{\partial}{\partial t}U(t,0) = -\mathrm{i}\,H(t)\,U(t,0), \tag{A.4}$$

analogous to (19) and the initial condition $U(0,0)=\mathbb{1}$. Moreover,

$$U(t-2\pi,-2\pi) = U(t,0), \tag{A.5}$$

due to the 2π-periodicity of $H(t)$.

Note that the special form of the Hamiltonian (7) due to circular polarization of the driving field implies

$$\overline{H(t)} = H(-t). \tag{A.6}$$

Define the family of unitaries $V(t,0)\equiv\overline{U(-t,0)}$. It satisfies

$$\frac{\partial}{\partial t}V(t,0) = -\overline{\frac{\partial}{\partial t}U(-t,0)} \overset{(A.4)}{=} -\overline{(-\mathrm{i}\,H(-t)U(-t,0))} \overset{(A.6)}{=} -\mathrm{i}\,H(t)\,V(t,0), \tag{A.7}$$

and $V(0,0)=\mathbb{1}$, the same differential equation and initial condition as $U(t,0)$. Hence

$$V(t,0) = U(t,0) = \overline{U(-t,0)} \quad \text{for all } t\in\mathbb{R}. \tag{A.8}$$

Especially, for $t=2\,\pi$,

$$U(2\pi,0) = \overline{U(-2\pi,0)} \overset{(A.3)}{=} \overline{U(0,-2\pi)^{-1}} \overset{(A.5)}{=} \overline{U(2\pi,0)^{-1}} = U(2\pi,0)^{\top}, \tag{A.9}$$

which completes the proof of $U(2\pi,0)$ being symmetric. □

Appendix B. Some properties of periodically driven *N*-level systems

We adopt a more general framework than in the main part of the paper and assume a Hamiltonian $H(\boldsymbol{\pi},t)$ as an Hermitean $N\times N$-matrix depending on certain parameters $\boldsymbol{\pi}\in\boldsymbol{\mathcal{P}}\subset\mathbb{R}^p$ including the driving frequency ω. Here the parameter space $\boldsymbol{\mathcal{P}}$ is assumed to be an open subset of $\mathbb{R}^p$. Again, the Hamiltonian will depend $T\equiv\frac{2\pi}{\omega}$-periodically on t. Moreover, we will assume that there exists a strictly monotone selection of quasienergies $\epsilon_n(\boldsymbol{\pi}),\ n=1,\ldots,N$ that depend smoothly on $\boldsymbol{\pi}\in\boldsymbol{\mathcal{P}}$:

Assumption B.1.

$$\epsilon_n(\boldsymbol{\pi}) < \epsilon_m(\boldsymbol{\pi}) \text{ for all } \ 1\leqslant n<m\leqslant N \text{ and } \boldsymbol{\pi}\in\boldsymbol{\mathcal{P}}. \tag{B.1}$$

Analogously to the definitions in section 4.2 we will define 'phases' $\boldsymbol{\mathcal{P}}_\nu\subset\boldsymbol{\mathcal{P}}$ by intersections of open subsets of $\boldsymbol{\mathcal{P}}$ of the form

$$\mathcal{O}^{>}_{nm\ell} \equiv \{\boldsymbol{\pi}\in\boldsymbol{\mathcal{P}}\,|\epsilon_n(\boldsymbol{\pi})-\epsilon_m(\boldsymbol{\pi})+\ell\omega>0\} \tag{B.2}$$

or

$$\mathcal{O}^{<}_{nm\ell} \equiv \{\boldsymbol{\pi}\in\boldsymbol{\mathcal{P}}\,|\epsilon_n(\boldsymbol{\pi})-\epsilon_m(\boldsymbol{\pi})+\ell\omega<0\}\,. \tag{B.3}$$

We are looking for 'minimal phases' in the sense that $\boldsymbol{\mathcal{P}}_\nu$ must not contain strictly smaller phases. Although the integer ℓ in (B.2) and (B.3) may assume infinitely many values it suffices to consider *finitely* many intersections of the above subsets. This can

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be seen as follows. Let $n>m$ such $\epsilon_n(\boldsymbol{\pi})-\epsilon_m(\boldsymbol{\pi})>0$. Then there exists an $\ell\in\mathbb{N}_0$ such that $\epsilon_n(\boldsymbol{\pi})-\epsilon_m(\boldsymbol{\pi})-\ell\,\omega>0$ but $\epsilon_n(\boldsymbol{\pi})-\epsilon_m(\boldsymbol{\pi})-(\ell+1)\,\omega<0$. It follows that for the pair (n,m) we need only consider the intersection of the two subsets $\mathcal{O}^{>}_{n,m,-\ell}$ and $\mathcal{O}^{<}_{n,m,-(\ell+1)}$ since the other ones of the form (B.2) or (B.3) are always larger and hence not minimal. Analogous considerations apply for the case $n<m$. It follows that the $\boldsymbol{\mathcal{P}}_\nu$ are open as finite intersections of open subsets of $\boldsymbol{\mathcal{P}}$.

The phase boundaries are again given by equations of the form

$$\epsilon_n(\boldsymbol{\pi})-\epsilon_m(\boldsymbol{\pi})+\ell\omega=0, \tag{B.4}$$

and will be denoted by $\boldsymbol{\mathcal{P}}_{nm\ell}$. It may happen, as in the case of the two spin Rabi model, that not all phase boundaries given by equations of the form (B.4) are realized since only a finite number of non-vanishing Fourier components of the relevant quantities exists.

Another problem is the requirement that the phase boundaries should have codimension one in $\boldsymbol{\mathcal{P}}$ whereas the 'avoided level crossing' of quasienergies, see, e.g. [39], is an indication of a larger codimension. To explain this problem in more detail we reconsider the $N\times N$ monodromy matrix $U(T,0)$ describing the unitary time evolution of the system after one period T and recall that the eigenvalues of $U(T,0)$ are in $1:1$ relation with equivalence classes of quasienergies modulo ω. A general unitary $N\times N$-matrix depends on N^2 real parameters, but the submanifold of unitary matrices with one pair of degenerate eigenvalues has only the dimension N^2-3, i.e. the codimension three. This supports the expectation that in the p-dimensional surface $\boldsymbol{\mathcal{P}}$ the phase boundaries given by (B.4) should also have codimension three, and not one as required in our approach. Note, however, that for special cases like the class of symmetric unitary matrices, see appendix A, the codimension reduces to two. Moreover, two eigenvalues of $U(T,0)$ belonging to different eigenvalues of a symmetry will not show the avoided level crossing, see, e.g. [39]. Another way to circumvent the above problem results when one of the parameters is the frequency of excitation ω. This frequency is constant for the monodromy matrix and the sketched argument for codimension three does not apply. As an illustration we remark that for the one spin $s=1/2$ Rabi problem with quasienergy $\epsilon_\pm=\frac{1}{2}(\omega\pm\Omega_{\mathrm{Rabi}})$, see (35), the crossing of quasienergies $\epsilon_+=\epsilon_-+\omega$ occurs for $\omega=\frac{f^2+\omega_0^2}{2\omega_0}$. The latter indicates a codimension one of the phase boundary in spite of the noncrossing rule.

The general definitions of section 4.1 also apply for the N level case. We note the following

Lemma B.1.

$$V^{(\ell)}_{nm}=\overline{V^{(-\ell)}_{mn}}\ \text{for all } n,m=1,\dots,N \text{ and } \ell\in\mathbb{Z}. \tag{B.5}$$

Proof. Recall that, due to V being Hermitean,

$$\tilde{V}_{nm}\stackrel{(61)}{=}\langle u_n(t)|\,V\,|u_m(t)\rangle=\sum_{\ell\in\mathbb{Z}}V^{(\ell)}_{nm}\,\mathrm{e}^{\mathrm{i}\,\ell\,\omega\,t}=\overline{\langle u_m(t)|\,V\,|u_n(t)\rangle}=\overline{\tilde{V}_{mn}}=\sum_{\ell\in\mathbb{Z}}\overline{V^{(\ell)}_{mn}}\,\mathrm{e}^{-\mathrm{i}\,\ell\,\omega\,t}=\sum_{\ell\in\mathbb{Z}}\overline{V^{(-\ell)}_{mn}}\,\mathrm{e}^{\mathrm{i}\,\ell\,\omega\,t}. \tag{B.6}$$

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The comparison of the coefficients of the first and the last Fourier series in (B.6) yields the result. □

Next we will formulate some arguments in favour of the following Assertion, albeit not in a mathematically rigorous manner.

Assertion 1. At least two NESS probabilities coincide at the phase boundaries.

Consider a fixed boundary $\boldsymbol{\mathcal{P}}_{\bar{n}\bar{m}\bar{\ell}}$ that is defined by the vanishing of some frequency $\omega^{(\bar{\ell})}_{\bar{m}\bar{n}}$. It follows from

$$\omega^{(\bar{\ell})}_{\bar{m}\bar{n}} = \epsilon_{\bar{m}} - \epsilon_{\bar{n}} + \bar{\ell}\omega = -\left(\epsilon_{\bar{n}} - \epsilon_{\bar{m}} - \bar{\ell}\omega\right) = -\omega^{(-\bar{\ell})}_{\bar{n}\bar{m}}, \tag{B.7}$$

see (62), that the complementary frequency $\omega^{(-\bar{\ell})}_{\bar{n}\bar{m}}$ vanishes too. For these values the thermal averages $N(\omega^{(\bar{\ell})}_{\bar{m}\bar{n}})$ and $N(\omega^{(-\bar{\ell})}_{\bar{n}\bar{m}})$ diverge due to (63). Hence close to the boundary these averages and the corresponding transition rates $\Gamma_{\bar{m}\bar{n}}$ and $\Gamma_{\bar{n}\bar{m}}$ will assume arbitrary large values. If the Pauli master equation (64) is written in the form

$$\sum_m \Gamma_{nm} p_m = \sum_m \Gamma_{mn} p_n, \tag{B.8}$$

it is obvious that for $n = \bar{n}$ both sides of (B.8) are dominated by a single term where $m = \bar{m}$ and hence

$$\Gamma_{\bar{n}\bar{m}} p_{\bar{m}} \approx \Gamma_{\bar{m}\bar{n}} p_{\bar{n}}. \tag{B.9}$$

This approximation is to be understood in the sense that although both sides of (B.9) become arbitrarily large its difference remains bounded. This means that close to the phase boundary we obtain a kind of 'local detailed balance' for the pair $(\bar{m}, \bar{n})$. On the other hand the matrix entries $\Gamma_{\bar{n}\bar{m}}$ will be almost symmetric, i.e. satisfy $\Gamma_{\bar{n}\bar{m}} \approx \Gamma_{\bar{m}\bar{n}}$ close to the phase boundary. This can be shown as follows. Using

$$\left|V^{(\bar{\ell})}_{\bar{m}\bar{n}}\right|^2 = \left|V^{(-\bar{\ell})}_{\bar{n}\bar{m}}\right|^2, \tag{B.10}$$

see lemma B.1 in this appendix, the limit relation

$$\lim_{\tilde{\omega}\downarrow 0} \frac{N(\tilde{\omega})}{N(-\tilde{\omega})} = \lim_{\tilde{\omega}\downarrow 0} \frac{1 - \mathrm{e}^{-\beta\tilde{\omega}}}{\mathrm{e}^{\beta\tilde{\omega}} - 1} = \lim_{\tilde{\omega}\downarrow 0} \mathrm{e}^{-\beta\,\tilde{\omega}} = 1, \tag{B.11}$$

and (B.7), we conclude

$$\Gamma_{\bar{m}\bar{n}} \approx \Gamma^{(\bar{\ell})}_{\bar{m}\bar{n}} \overset{(60)}{=} 2\,\pi \left|V^{(\bar{\ell})}_{\bar{m}\bar{n}}\right|^2 N(\omega^{(\bar{\ell})}_{\bar{m}\bar{n}})\, J_0 \approx 2\,\pi \left|V^{(-\bar{\ell})}_{\bar{n}\bar{m}}\right|^2 N(\omega^{(-\bar{\ell})}_{\bar{n}\bar{m}})\, J_0 = \Gamma^{(-\bar{\ell})}_{\bar{n}\bar{m}} \approx \Gamma_{\bar{n}\bar{m}}. \tag{B.12}$$

Consequently, when approaching the phase boundary, symbolically denoted by $\lim_{\tilde{\omega}\downarrow 0}$, we have

$$\lim_{\tilde{\omega}\downarrow 0} \frac{p_{\bar{m}}}{p_{\bar{n}}} \overset{(B.9)}{=} \lim_{\tilde{\omega}\downarrow 0} \frac{\Gamma_{\bar{m}\bar{n}}}{\Gamma_{\bar{n}\bar{m}}} \overset{(B.12)}{=} 1, \tag{B.13}$$

which completes the arguments in favour of assertion 1. □

In the case of a single spin s all quasienergy levels are equidistant, see equations (53) and (54) in [29], and thus the coincidence of two probabilities at the phase boundary

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implies that all probabilities p_n are the same and hence the inverse quasitemperature vanishes, see [29].

In the general case arguments analogous to those at the end of section 4.2 show that the NESS will not be of Boltzmann type at least at the phase boundaries and, by continuity, in a small neighbourhood of the phase boundaries. This supports the conjecture that the existence of a quasitemperature of the NESS is restricted to very special systems.

Next we will address the question how the NESS probabilities p_n are connected at the phase boundaries and formulate the following

Assertion 2. The NESS probabilities are continuous at the phase boundaries but their gradients are discontinuous there.

We will provide some arguments in favour of this assertion that could probably be strengthen to a more rigorous proof. To this end we consider a fixed phase boundary $\boldsymbol{\mathcal{P}}_{\bar{n}\bar{m}\bar{\ell}}$ given by the equation

$$0 = \omega_{\bar{n}\bar{m}}^{(\bar{\ell})} = \epsilon_{\bar{n}} - \epsilon_{\bar{m}} + \bar{\ell}\,\omega, \tag{B.14}$$

and will calculate the p_n in a small neighbourhood of some point $\boldsymbol{\pi} \in \boldsymbol{\mathcal{P}}_{\bar{n}\bar{m}\bar{\ell}}$. We consider a curve through $\boldsymbol{\pi}$ perpendicular to $\boldsymbol{\mathcal{P}}_{\bar{n}\bar{m}\bar{\ell}}$ parametrized by the parameter

$$x \equiv \beta\,\omega_{\bar{n}\bar{m}}^{(\bar{\ell})}, \tag{B.15}$$

such that $-\delta < x < \delta$ for some $\delta > 0$ and $x = 0$ corresponds to the point $\boldsymbol{\pi} \in \boldsymbol{\mathcal{P}}_{\bar{n}\bar{m}\bar{\ell}}$.

First we only consider the 'positive neighbourhood' $\boldsymbol{\mathcal{P}}^{>}_{\bar{n}\bar{m}\bar{\ell}}$ of $\boldsymbol{\mathcal{P}}_{\bar{n}\bar{m}\bar{\ell}}$ given by $\omega_{\bar{n}\bar{m}}^{(\bar{\ell})} > 0$ (such that also $x > 0$) and restricted in such a way that no other phase boundaries intersect $\boldsymbol{\mathcal{P}}^{>}_{\bar{n}\bar{m}\bar{\ell}}$. We assume that a Taylor series representation of p_n holds in $\boldsymbol{\mathcal{P}}^{>}_{\bar{n}\bar{m}\bar{\ell}}$ with the first terms being of the form

$$p_n = p_{n0} + x\,p_{n1} + O(x^2). \tag{B.16}$$

We denote by $\Gamma^{>}$ and $\widetilde{\Gamma}^{>}$ the transition rate matrix functions (59) and (65) restricted to the positive neighbourhood $\boldsymbol{\mathcal{P}}^{>}_{\bar{n}\bar{m}\bar{\ell}}$. According to what has been said the matrix entries $\Gamma^{>}_{nm}$ will be smooth functions of x for $-\delta < x < \delta$ except for $\Gamma^{>}_{\bar{n}\bar{m}}$ and $\Gamma^{>}_{\bar{m}\bar{n}}$ where the transition rates diverge for $x \to 0$. Hence it is sensible to adopt Laurent series representations for the $\Gamma^{>}_{nm}$ that are Taylor series for most cases but start with an $\frac{1}{x}$-term in the latter two cases.

In particular, isolating the diverging terms, we may write

$$\widetilde{\Gamma}^{>}_{\bar{n}\bar{m}} = \Gamma^{>}_{\bar{n}\bar{m}} = 2\pi J_0 \left\{ \left| V_{\bar{n}\bar{m}}^{(\bar{\ell})} \right|^2 \frac{1}{\mathrm{e}^{x} - 1} + \sum_{\substack{\ell \in \mathbb{Z} \\ \ell \neq \bar{\ell}}} \left| V_{\bar{n}\bar{m}}^{(\ell)} \right|^2 N\left(\omega_{\bar{n}\bar{m}}^{\ell}\right) \right\}, \tag{B.17}$$

and

$$\widetilde{\Gamma}^{>}_{\bar{m}\bar{n}} = \Gamma^{>}_{\bar{m}\bar{n}} = 2\pi J_0 \left\{ \left| V^{(-\bar{\ell})}_{\bar{m}\bar{n}} \right|^2 \frac{1}{1-\mathrm{e}^{-x}} + \sum_{\substack{\ell\in\mathbb{Z} \\ \ell\neq -bar\ell}} \left| V^{(\ell)}_{\bar{m}\bar{n}} \right|^2 N\left(\omega^{\ell}_{\bar{m}\bar{n}}\right) \right\}. \tag{B.18}$$

For the modified matrix $\widetilde{\Gamma}^{>}$ additionally two diagonal elements will diverge for $x \to 0$. According to

$$\widetilde{\Gamma}^{>}_{\bar{m}\bar{m}} = \Gamma^{>}_{\bar{m}\bar{m}} - \sum_n \Gamma^{>}_{n\bar{m}}, \tag{B.19}$$

see (65), the diverging term of $\widetilde{\Gamma}^{>}_{\bar{m}\bar{m}}$ is

$$-2\pi\, J_0 \left| V^{(\bar{\ell})}_{\bar{n}\bar{m}} \right|^2 \frac{1}{\mathrm{e}^{x}-1}. \tag{B.20}$$

Analogously, the diverging term of $\widetilde{\Gamma}^{>}_{\bar{n}\bar{n}}$ is

$$-2\pi\, J_0 \left| V^{(-\bar{\ell})}_{\bar{m}\bar{n}} \right|^2 \frac{1}{1-\mathrm{e}^{-x}}. \tag{B.21}$$

All terms in (B.16)–(B.21) can be written as Taylor series in x with the exception of the highlighted exponential terms that possess the Laurent series

$$\frac{1}{\mathrm{e}^{x}-1} = \frac{1}{x} - \frac{1}{2} + \frac{x}{12} + O(x^2), \tag{B.22}$$

and

$$\frac{1}{1-\mathrm{e}^{-x}} = \frac{1}{x} + \frac{1}{2} + \frac{x}{12} + O(x^2). \tag{B.23}$$

Recall that the vector $\mathbf{p}^{>}$ of NESS probabilities in the positive neighbourhood is the (normalized) solution of $\widetilde{\Gamma}^{>}\mathbf{p}^{>} = 0$ that is unique due to theorem 2. After expanding $\widetilde{\Gamma}^{>}$ and $\mathbf{p}^{>}$ into Laurent series w.r.t. x we will set the first three coefficients of the resulting Laurent series of $\widetilde{\Gamma}^{>}\mathbf{p}^{>}$ to zero and thus obtain the first two terms of (B.16). These will determine the limit of the NESS probabilities and its gradient at the phase boundary.

In order to keep the representation as simple as possible we will, without loss of generality, assume that $\bar{n} = 1$ and $\bar{m} = 2$. It will suffice to give the structure of the Laurent series of $\widetilde{\Gamma}^{>}$ without going into the details of how the various numbers can be expressed by the physical quantities:

$$\widetilde{\Gamma}^{>} = \begin{pmatrix} -\frac{a}{x} + d + \dots & \frac{a}{x} + b + \dots & \mathbf{a}_0^\top + x\,\mathbf{a}_1^\top \\ \frac{a}{x} + c + \dots & -\frac{a}{x} + e + \dots & \mathbf{c}_0^\top + x\,\mathbf{c}_1^\top \\ \mathbf{b}_0 + x\,\mathbf{b}_1 & \mathbf{d}_0 + x\,\mathbf{d}_1 & \boldsymbol{\gamma}_0 + x\,\boldsymbol{\gamma}_1 \end{pmatrix} + O(x^2). \tag{B.24}$$

Here we have omitted the x-linear terms in the upper left 2×2-submatrix that are not needed in the sequel. The real numbers a, b, c, d, e are independent of x, likewise the $(N-2)$-dimensional vectors $\mathbf{a}_0, \dots, \mathbf{d}_1$ and the $(N-2)\times(N-2)$-matrices $\boldsymbol{\gamma}_0$ and $\boldsymbol{\gamma}_1$. We stress that the repeated occurrence of the quantity

$$a = 2\pi\, J_0 \lim_{x\downarrow 0} \left| V^{(\bar{\ell})}_{12} \right|^2 \tag{B.25}$$

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in (B.24) is crucial for the following considerations. The vector of NESS probabilities $\mathbf{p}^{>}$ will be written as

$$\mathbf{p}^{>} = \begin{pmatrix} p_{10} + x\, p_{11} \\ p_{20} + x\, p_{21} \\ \mathbf{p}_0 + x\, \mathbf{p}_1 \end{pmatrix} + O(x^2). \tag{B.26}$$

Setting the coefficients of the resulting Laurent series of the various components of $\widetilde{\Gamma}^{>}\mathbf{p}^{>}$ to zero yields the following results:

$$x^{-1} : \frac{a}{x}\,(p_{20} - p_{10}) = 0 \;\Rightarrow\; p_{20} = p_{10} \equiv p, \tag{B.27}$$

$$x^{0} : p\,(\mathbf{b}_0 + \mathbf{d}_0) + \boldsymbol{\gamma}_0\, \mathbf{p}_0 = 0 \;\Rightarrow\; \mathbf{p}_0 = -p\, \boldsymbol{\gamma}_0^{-1}\,(\mathbf{b}_0 + \mathbf{d}_0)\,, \tag{B.28}$$

$$x^{0} : a \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix} \begin{pmatrix} p_{11} \\ p_{21} \end{pmatrix} = \begin{pmatrix} -p(d+b) - \mathbf{a}_0 \cdot \mathbf{p}_0 \\ -p(c+e) - \mathbf{c}_0 \cdot \mathbf{p}_0 \end{pmatrix} \tag{B.29}$$

$$\Rightarrow\; p_{11} = -\frac{1}{2a}\,(p(d+b+c+e) + \mathbf{a}_0 \cdot \mathbf{p}_0 + \mathbf{c}_0 \cdot \mathbf{p}_0) \text{ and} \tag{B.30}$$

$$p_{21} = -\frac{1}{2a}\,(p(d+b-c-e) + \mathbf{a}_0 \cdot \mathbf{p}_0 - \mathbf{c}_0 \cdot \mathbf{p}_0)\,, \tag{B.31}$$

$$x^{1} : x\,(p_{11}\, \mathbf{b}_0 + p\, \mathbf{b}_1 + p_{21}\, \mathbf{d}_0 + p\, \mathbf{d}_1 + \boldsymbol{\gamma}_1\, \mathbf{p}_0 + \boldsymbol{\gamma}_0\, \mathbf{p}_1) = 0 \tag{B.32}$$

$$\Rightarrow\; \mathbf{p}_1 = -\boldsymbol{\gamma}_0^{-1}\,(-\boldsymbol{\gamma}_1\, \mathbf{p}_0 + p\,(\mathbf{b}_1 + \mathbf{d}_1) + p_{11}\, \mathbf{b}_0 + p_{21}\, \mathbf{d}_0)\,. \tag{B.33}$$

A few remarks are in order. First, we note that the result $p_{20} = p_{10} \equiv p$ in (B.27) again confirms the previous statement in assertion 1 that at least two NESS probabilities coincide at the phase boundaries. Of course, the free parameter $p > 0$ has to be chosen in such a way that the probabilities sum up to unity.

Second, we have used in (B.28) and (B.33) that $\boldsymbol{\gamma}_0$ is invertible. This can be shown as follows. Let, for $-\delta < x < \delta$, $\Gamma^{\wedge}(x)$ denote the matrix obtained from $\Gamma^{>}(x)$ by subtracting its principle part, i.e. the terms of the form $\pm\frac{a}{x}$, analogously for $\widetilde{\Gamma}^{\wedge}(x)$. Then it can be easily shown that $\Gamma^{\wedge}(x)$ also satisfies the conditions of theorem 2. Hence $\widetilde{\Gamma}^{\wedge}(x)$ has an one-dimensional null space spanned by some $p^{\wedge} > 0$. This vector cannot lie in the subspace of vectors of the form $(0, 0, \mathbf{p})^{\top}$ and the matrix $\boldsymbol{\gamma}(x)$, defined as the restriction of $\widetilde{\Gamma}^{\wedge}(x)$ to this subspace, must be invertible for all $-\delta < x < \delta$. Especially, $\boldsymbol{\gamma}_0 = \boldsymbol{\gamma}(0)$ is invertible.

The calculations with $\widetilde{\Gamma}^{<}$ and $p^{<}$ defined in the 'negative neighbourhood' $\boldsymbol{\mathcal{P}}^{<}_{\bar{n}\bar{m}\bar{\ell}}$ of $\boldsymbol{\mathcal{P}}_{\bar{n}\bar{m}\bar{\ell}}$ given by $\omega^{(\bar{\ell})}_{\bar{n}\bar{m}} < 0$ are completely analogous and need not be given in detail. The only difference is that for $x < 0$ we have

$$N(\omega^{(\ell)}_{12}) = \frac{1}{1 - \mathrm{e}^{x}} = -\frac{1}{x} + \frac{1}{2} - \frac{x}{12} + O(x^2), \tag{B.34}$$

and

$$N(\omega_{21}^{(-\ell)}) = \frac{1}{e^{-x} - 1} = -\frac{1}{x} - \frac{1}{2} - \frac{x}{12} + O(x^2). \tag{B.35}$$

This means that the Laurent series for $\widetilde{\Gamma}^<$ is identical with (B.24), with the only exception that a has to be replaced by $-a$. This modification does not change the solution for $p_{10} = p_{20} = p$ according to (B.27) and for $\mathbf{p}_0$ according to (B.28). Hence the NESS probabilities are continuous at the phase boundaries. In contrast, the solutions for p_{11} and p_{21} according to (B.29) and (B.31) will change their sign and hence also $\mathbf{p}_1$ according to (B.33) will be different for the negative neighbourhood. This means that the x-derivative and hence the gradient of the NESS probabilities will be discontinuous at the phase boundaries, thereby completing the arguments in favour of assertion 2. $\square$

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