Periodic thermodynamics of the Rabi model with circular polarization for arbitrary spin quantum numbers

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We consider a spin s subjected to both a static and an orthogonally applied oscillating, circularly polarized magnetic field while being coupled to a heat bath, and analytically determine the quasi-stationary distribution of its Floquet-state occupation probabilities for arbitrarily strong driving. This distribution is shown to be Boltzmannian with a quasitemperature which is different from the temperature of the bath, and independent of the spin quantum number. We discover a remarkable formal analogy between the quasithermal magnetism of the nonequilibrium steady state of a driven ideal paramagnetic material, and the usual thermal paramagnetism. Nonetheless, the response of such a material to the combined fields is predicted to show several unexpected features, even allowing one to turn a paramagnet into a diamagnet under strong driving. Thus, we argue that experimental measurements of this response may provide key paradigms for the emerging field of periodic thermodynamics.

I. INTRODUCTION

A quantum system governed by an explicitly time-dependent Hamiltonian $H(t)$ which depends *periodically* on 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

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

The *Floquet functions* $|u_n(t)\rangle$ share the T-periodic time dependence of their Hamiltonian,

$$|u_n(t)\rangle = |u_n(t+T)\rangle \; ; \tag{3}$$

the quantities ε_n, which accompany their time evolution in the same manner as energy eigenvalues accompany the evolution of unperturbed energy eigenstates, are known as *quasienergies* [1–3]. Here we assume that the quasienergies constitute a pure point spectrum, associated with square-integrable Floquet states in the system's Hilbert space $\mathcal{H}_S$; we also adopt a system of units such that both the Planck constant $\hbar$ and the Boltzmann constant k_{B} are set to one.

Evidently the factorization of a Floquet state (2) into a Floquet function and an exponential of a phase which grows linearly in time is not unique: Defining $\omega = 2\pi/T$, and taking an arbitrary, positive or negative integer ν, one has

$$|u_n(t)\rangle \exp(-\mathrm{i}\varepsilon_n t) = |u_n(t)\mathrm{e}^{\mathrm{i}\nu\omega t}\rangle \exp\bigl(-\mathrm{i}[\varepsilon_n+\nu\omega]t\bigr) \; , \tag{4}$$

where $|u_n(t)\mathrm{e}^{\mathrm{i}\nu\omega t}\rangle$ again is a T-periodic Floquet function, representing the same Floquet state as $|u_n(t)\rangle$. Therefore, a quasienergy is not to be regarded as just a single number equipped with the dimension of energy, but rather as an infinite set of equivalent representatives,

$$\varepsilon_n \equiv \{\varepsilon_n + \nu\omega \mid \nu \in \mathbb{Z}\} \; , \tag{5}$$

where the choice of the "canonical representative" distinguished by setting $\nu = 0$ is a matter of convention.

The significance of these Floquet states (2) rests in the fact that, as long as the Hamiltonian depends on time in a strictly T-periodic manner, every solution $|\psi(t)\rangle$ to the time-dependent Schrödinger equation can be expanded with respect to the Floquet basis,

$$|\psi(t)\rangle = \sum_n c_n \, |u_n(t)\rangle \exp(-\mathrm{i}\varepsilon_n t) \; , \tag{6}$$

where 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. Under conditions of perfectly coherent time evolution these coefficients c_n would be determined solely by the system's state at the moment the periodic drive is turned on. 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, to the effect that a quasistationary distribution $\{p_n\}$ of Floquet-state occupation probabilities establishes itself which contains no memory of the initial state, and the question emerges how to quantify this distribution.

In a short programmatic note entitled "Periodic Thermodynamics", Kohn has drawn attention to such quasistationary Floquet-state distributions $\{p_n\}$, emphasizing that they should be less universal than usual distributions characterizing thermal equilibrium, depending on the very form of the system's interaction with its environment [10]. 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]. For the particular case of a linearly forced *harmonic* oscillator these authors have shown that the Floquet-state distribution remains a Boltzmann distribution with the temperature of the heat bath, whereas it becomes rather more complicated in the case of forced *anharmonic* oscillators. These investigations have been extended later by Ketzmerick and Wustmann, who have demonstrated that structures found in the phase space of classical forced anharmonic oscillators leave their distinct traces in the quasistationary Floquet-state distributions of their quantum counterparts [12]. To date, a great variety of different individual aspects of the "periodic thermodynamics" envisioned by Kohn has been discussed in the literature [13–23], 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 and thus to unravel salient features on the one hand, and which actually open up meaningful perspectives for new laboratory experiments on the other. To this end, in the present work we consider 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 [24], and coupled to a thermal bath of harmonic oscillators. The experimental measurement of the thermal paramagnetism resulting from magnetic moments subjected to a static field alone has a long and successful history [25, 26], having become a standard topic in textbooks on Statistical Physics [27, 28]. We argue that a future generation of such experiments, including both a static and a strong oscillating field, may set further cornerstones towards the development of full-fledged periodic thermodynamics.

We proceed as follows: In Sec. II we collect the necessary technical tools, starting with a brief summary of the golden-rule approach to time-periodically driven open quantum systems in the form developed by Breuer *et al.* [11], thereby establishing our notation. We also sketch a technique which enables one to "lift" a solution to the Schrödinger equation for a spin $s = \frac{1}{2}$ in a time-varying magnetic field to general s. In Sec. III we discuss the Floquet states for spins in a circularly polarized driving field, obtaining the states for general s from those for $s = \frac{1}{2}$ with the help of the lifting procedure. In Sec. IV we compute the quasistationary Floquet-state distribution for driven spins under the assumption that the spectral density of the heat bath be constant, and show that this distribution is Boltzmannian with a quasitemperature which is *different* from the actual bath temperature; the dependence of this quasitemperature on the system parameters is discussed in some detail. In Sec. V we determine the magnetization of a spin system which is subjected to both a static and an orthogonally applied, circularly polarized magnetic field while being coupled to a heat bath. To this end, we first establish a general formula for the ensuing magnetization by means of another systematic use of the lifting technique, and then show that the resulting expression can be interpreted as a derivative of a partition function based on both the quasitemperature and the system's quasienergies, in perfect formal analogy to the textbook treatment of paramagnetism in the absence of time-periodic driving; these insights are exploited for elucidating the response of an ideal paramagnet to a circularly polarized driving field. In Sec. VI we consider the rate of energy dissipated by the driven spins into the bath, thus generalizing results derived previously for $s = \frac{1}{2}$ in Ref. [29]. In Sec. VII we summarize and discuss our main findings, emphasizing the possible knowledge gain to be derived from future measurements of paramagnetic response to strong time-periodic forcing, carried out along the lines drawn in the present work.

II. TECHNICAL TOOLS

A. Golden-rule approach to open driven systems

Let us consider a quantum system evolving according to a T-periodic Hamiltonian $H(t)$ on a Hilbert space $\mathcal{H}_S$ which is perturbed by a time-independent operator V. Then the transition matrix element connecting an initial Floquet function $|u_i(t)\rangle$ to a final Floquet function $|u_f(t)\rangle$ can be expanded into a Fourier series,

$$\langle u_f(t)| V |u_i(t)\rangle = \sum_{\ell\in\mathbb{Z}} V_{fi}^{(\ell)} \exp(\mathrm{i}\ell\omega t) \; , \tag{7}$$

and consequently the "golden rule" for the rate of transitions Γ_{fi} from a Floquet state labeled i to a Floquet state f is written as [29]

$$\Gamma_{fi} = 2\pi \sum_{\ell\in\mathbb{Z}} |V_{fi}^{(\ell)}|^2 \, \delta(\omega_{fi}^{(\ell)}) \; , \tag{8}$$

where

$$\omega_{fi}^{(\ell)} = \varepsilon_f - \varepsilon_i + \ell\omega \; . \tag{9}$$

Thus, a transition among Floquet states is not 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 (5); this is one of the sources of the peculiarities which distinguish periodic thermodynamics from usual equilibrium thermodynamics [10, 11].

Let us now assume that, instead of merely being perturbed by V, the periodically driven system is coupled to a heat bath, described by a Hamiltonian H_{bath} acting on a Hilbert space $\mathcal{H}_B$, so that the total Hamiltonian on the composite Hilbert space $\mathcal{H}_S \otimes \mathcal{H}_B$ takes the form

$$H_{\mathrm{total}}(t) = H(t) \otimes \mathbb{1} + \mathbb{1} \otimes H_{\mathrm{bath}} + H_{\mathrm{int}} \; . \tag{10}$$

Stipulating further that the interaction Hamiltonian H_{int} factorizes according to

$$H_{\mathrm{int}} = V \otimes W \; , \tag{11}$$

the golden rule can be applied to joint transitions from Floquet states i to Floquet states f of the system accompanied by transitions from bath eigenstates n with energy E_n to other bath eigenstates m with energy E_m, acquiring the form

$$\Gamma_{fi}^{mn} = 2\pi \sum_{\ell \in \mathbb{Z}} |V_{fi}^{(\ell)}|^2 \, |W_{mn}|^2 \, \delta(E_m - E_n + \omega_{fi}^{(\ell)}) \; . \quad (12)$$

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

$$W = \sum_{\widetilde{\omega}} \left(b_{\widetilde{\omega}} + b_{\widetilde{\omega}}^\dagger \right) \; , \quad (13)$$

where $b_{\widetilde{\omega}}$ ($b_{\widetilde{\omega}}^\dagger$) is the annihilation (creation) operator pertaining to a bath oscillator of frequency $\widetilde{\omega}$. We now have to distinguish two cases: If $E_n - E_m = \widetilde{\omega} > 0$, so that the bath is de-excited and transfers energy to the system, the required annihilation-operator matrix element reads

$$W_{mn} = \sqrt{n(\widetilde{\omega})} \; , \quad (14)$$

where $n(\widetilde{\omega})$ is the occupation number of a bath oscillator with frequency $\widetilde{\omega}$, and the square $|W_{mn}|^2$ entering the golden rule (12) has to be replaced by the thermal avarage

$$N(\widetilde{\omega}) \equiv \langle n(\widetilde{\omega}) \rangle = \frac{1}{\exp(\beta\widetilde{\omega}) - 1} \; , \quad (15)$$

with β denoting the inverse bath temperature. Conversely, if $E_n - E_m = \widetilde{\omega} < 0$ so that the system loses energy to the bath and a bath phonon is created, one has

$$W_{mn} = \sqrt{n(-\widetilde{\omega}) + 1} \; , \quad (16)$$

giving

$$N(\widetilde{\omega}) \equiv \langle n(-\widetilde{\omega}) \rangle + 1 = \frac{1}{1 - \exp(\beta\widetilde{\omega})} \; . \quad (17)$$

Finally, let $J(\widetilde{\omega})$ be the spectral density of the bath. Then the total rate Γ_{fi} of bath-induced transitions among the Floquet states i and f of the driven system is expressed as a sum of partial rates,

$$\Gamma_{fi} = \sum_{\ell \in \mathbb{Z}} \Gamma_{fi}^{(\ell)} \; , \quad (18)$$

where

$$\Gamma_{fi}^{(\ell)} = 2\pi \, |V_{fi}^{(\ell)}|^2 \, N(\omega_{fi}^{(\ell)}) \, J(|\omega_{fi}^{(\ell)}|) \; . \quad (19)$$

These total rates (18) now determine the desired quasistationary distribution $\{p_n\}$ as a solution to the equation [11]

$$\sum_m \left(\Gamma_{nm} p_m - \Gamma_{mn} p_n \right) = 0 \; . \quad (20)$$

It deserves to be emphasized again that the very details of the system-bath coupling enter here, so that the precise form of the respective distribution $\{p_n\}$ may depend strongly on such details [10].

B. The lift from $s = \frac{1}{2}$ to general s

We will make heavy use of a procedure which allows one to transfer a solution to the Schrödinger equation for a spin with spin quantum number $s = \frac{1}{2}$ in a time-dependent external field to a solution of the corresponding Schrödinger equation for general s, see also Ref. [30]. This procedure does not appear to be widely known, but has been applied already in 1987 to the coherent evolution of a laser-driven N-level system possessing an $SU(2)$ dynamic symmetry [31], and more recently to the spin-s Landau-Zener problem [32]. Here we briefly sketch this method.

Let $t \mapsto \Psi(t) \in SU(2)$ be a smooth curve such that $\Psi(0) = \mathbb{1}$, as given by a 2×2-matrix of the form

$$\Psi(t) = \begin{pmatrix} z_1(t) & z_2(t) \\ -z_2^*(t) & z_1^*(t) \end{pmatrix} \quad (21)$$

with complex functions $z_1(t)$, $z_2(t)$ obeying $|z_1(t)|^2 + |z_2(t)|^2 = 1$ for all times t. One then has

$$\left(\frac{\mathrm{d}}{\mathrm{d}t} \Psi(t) \right) \Psi(t)^{-1} \equiv -\mathrm{i} H(t) \in su(2) \; , \quad (22)$$

where $su(2)$ denotes the Lie algebra of $SU(2)$, *i.e.*, the space of anti-Hermitean, traceless 2×2-matrices which is closed under commutation [33]. Hence the columns $|\psi_1(t)\rangle$, $|\psi_2(t)\rangle$ of $\Psi(t)$ are linearly independent solutions of the Schrödinger equation

$$\mathrm{i} \frac{\mathrm{d}}{\mathrm{d}t} |\psi_j(t)\rangle = H(t) \, |\psi_j(t)\rangle \; , \quad j = 1, 2 \; . \quad (23)$$

Next we consider the well-known irreducible Lie algebra representation of $su(2)$,

$$r^{(s)} : su(2) \longrightarrow su(2s+1) \; , \quad (24)$$

which is parametrized by a spin quantum number s such that $2s \in \mathbb{N}$, together with the corresponding irreducible group representation (“irrep” for brevity)

$$R^{(s)} : SU(2) \longrightarrow SU(2s+1) \; . \quad (25)$$

It follows that

$$r^{(s)}(\mathrm{i}s_j) = \mathrm{i}S_j \; , \quad j = x, y, z \; , \quad (26)$$

where $s_j = \frac{1}{2}\sigma_j$ denote the three $s = \frac{1}{2}$ spin operators given by the Pauli matrices σ_j, and the S_j denote the corresponding spin operators for general s. Recall the standard matrices

$$\begin{aligned} (S_z)_{m,n} &= n \, \delta_{mn} \; , \qquad (27) \\ (S_x)_{m,n} &= \begin{cases} \frac{1}{2}\sqrt{s(s+1) - n(n \pm 1)} & : \; m = n \pm 1 \; , \\ 0 & : \; \text{else} \; , \end{cases} \\ (S_y)_{m,n} &= \begin{cases} \pm\frac{1}{2\mathrm{i}}\sqrt{s(s+1) - n(n \pm 1)} & : \; m = n \pm 1 \; , \\ 0 & : \; \text{else} \; , \end{cases} \end{aligned}$$

where $m, n = s, s-1, \ldots, -s$, and

$$S_\pm \equiv S_x \pm \mathrm{i} S_y \; . \qquad (28)$$

It follows from the general theory of representations [33] that $r^{(s)}$ and $R^{(s)}$ may be applied to Eq. (22) and yield

$$\left(\frac{\mathrm{d}}{\mathrm{d}t} R^{(s)}\, \Psi(t) \right) \Big(R^{(s)}\, \Psi(t) \Big)^{-1} = r^{(s)}\big(-\mathrm{i}H(t) \big) \; . \qquad (29)$$

Since $H(t)$ can always be written in the form of a Zeeman term with a time-dependent magnetic field $\mathbf{b}(t)$, namely,

$$H(t) = \mathbf{b}(t) \cdot \mathbf{s} = \sum_{j=1}^{3} b_j(t)\, s_j \; , \qquad (30)$$

Eq. (26) implies that

$$r^{(s)}\big(\mathrm{i}H(t) \big) = \mathrm{i}\, \mathbf{b}(t) \cdot \mathbf{S} = \mathrm{i} \sum_{j=1}^{3} b_j(t)\, S_j \; . \qquad (31)$$

In (30) we have chosen the $+$ sign convention adapted to electrons for which the magnetic moment is opposite to their spin $\mathbf{s}$. Hence, the "lifted" matrix

$$\Psi^{(s)}(t) \equiv R^{(s)}\big(\Psi(t) \big) \qquad (32)$$

will be a matrix solution of the lifted Schrödinger equation

$$\mathrm{i} \frac{\mathrm{d}}{\mathrm{d}t} \Psi^{(s)}(t) = \mathbf{b}(t) \cdot \mathbf{S}\, \Psi^{(s)}(t) \; . \qquad (33)$$

Note that the matrix $\Psi^{(s)}(t)$ is unitary, and hence its columns span the general $(2s+1)$-dimensional solution space of the lifted Schrödinger equation (33).

III. FLOQUET FORMULATION OF THE RABI PROBLEM

A. Floquet decomposition for $s = 1/2$

A spin $\frac{1}{2}$ subjected to both a constant magnetic field applied in the z-direction and an orthogonal, circularly polarized time-periodic field, as constituting the classic Rabi problem [24], is described by the Hamiltonian

$$H(t) = \frac{\omega_0}{2} \sigma_z + \frac{F}{2} (\sigma_x \cos\omega t + \sigma_y \sin\omega t) \; . \qquad (34)$$

Here ω_0 denotes the transition frequency pertaining to the spin states in the static field alone, while F denotes the frequency associated with the amplitude of the periodic drive. This is a special form of the Zeeman Hamiltonian (30) with the particular choices

$$\begin{aligned} b_x(t) &= F \cos\omega t \\ b_y(t) &= F \sin\omega t \\ b_z(t) &= \omega_0 \; . \end{aligned} \qquad (35)$$

The Floquet states (2) brought about by this Hamiltonian (34) are given by [34]

$$|\psi_\pm(t)\rangle = \frac{\mathrm{e}^{\mp \mathrm{i}\Omega t/2}}{\sqrt{2\Omega}} \begin{pmatrix} \pm\sqrt{\Omega \pm \delta}\; \mathrm{e}^{-\mathrm{i}\omega t/2} \\ \sqrt{\Omega \mp \delta}\; \mathrm{e}^{+\mathrm{i}\omega t/2} \end{pmatrix} \; , \qquad (36)$$

where

$$\delta = \omega_0 - \omega \qquad (37)$$

denotes the detuning of the transition frequency ω_0 from the driving frequency ω, and Ω is the Rabi frequency,

$$\Omega = \sqrt{\delta^2 + F^2} \; . \qquad (38)$$

The 2×2-matrix $\Psi(t)$ constructed from these states does not satisfy $\Psi(0) = \mathbb{1}$. This is of no concern, since $\Psi(t)$ could be replaced by $\Psi(t)\big(\Psi(0)\big)^{-1}$. The distinct advantage of these Floquet solutions (36) lies in the fact that they yield a particularly convenient starting point for the lifting procedure outlined in Sec. II B: One has

$$\begin{aligned} \Psi(t) &= \frac{1}{\sqrt{2\Omega}} \begin{pmatrix} \sqrt{\Omega+\delta} & -\sqrt{\Omega-\delta} \\ \mathrm{e}^{\mathrm{i}\omega t}\sqrt{\Omega-\delta} & \mathrm{e}^{\mathrm{i}\omega t}\sqrt{\Omega+\delta} \end{pmatrix} \\ &\quad \times \begin{pmatrix} \mathrm{e}^{-\mathrm{i}(\omega+\Omega)t/2} & 0 \\ 0 & \mathrm{e}^{-\mathrm{i}(\omega-\Omega)t/2} \end{pmatrix} \\ &\equiv P(t)\, \mathrm{e}^{-\mathrm{i}\omega t/2} \exp(-\mathrm{i}\Omega t\, s_z) \; . \end{aligned} \qquad (39)$$

This decomposition possesses the general Floquet form

$$\Psi(t) = P(t) \exp(-\mathrm{i}Gt) \; , \qquad (40)$$

where the unitary matrix $P(t) = P(t+T)$ again is T-periodic, and the eigenvalues of the "Floquet matrix" G, to be obtained from the matrix logarithm of $\Psi(T)\big(\Psi(0)\big)^{-1} = \exp(-\mathrm{i}GT)$, provide the system's quasienergies [35–38]. Since G already is diagonal in this representation (39), the quasienergies of a spin $\frac{1}{2}$ driven by a circularly polarized field according to the Hamiltonian (34) can be read off immediately:

$$\varepsilon_\pm = \frac{\omega \pm \Omega}{2} \mod \omega \; , \qquad (41)$$

satisfying $\varepsilon_+ + \varepsilon_- = 0 \mod \omega$. For later application we express the periodic part $P(t)$ of the decomposition (39) in the following way:

$$\begin{aligned} P(t) &= \mathrm{e}^{\mathrm{i}\omega t/2} \begin{pmatrix} \mathrm{e}^{-\mathrm{i}\omega t/2} & 0 \\ 0 & \mathrm{e}^{+\mathrm{i}\omega t/2} \end{pmatrix} \\ &\quad \times \frac{1}{\sqrt{2\Omega}} \begin{pmatrix} \sqrt{\Omega+\delta} & -\sqrt{\Omega-\delta} \\ \sqrt{\Omega-\delta} & \sqrt{\Omega+\delta} \end{pmatrix} \\ &\equiv \mathrm{e}^{\mathrm{i}\omega t/2} \exp(-\mathrm{i}\omega t\, s_z)\, \Xi \; . \end{aligned} \qquad (42)$$

The time-independent matrix $\Xi = \Psi(0)$ introduced here can be written as

$$\Xi = \exp\left(-\mathrm{i}\lambda\, s_y\right) = \begin{pmatrix} \cos(\lambda/2) & -\sin(\lambda/2) \\ \sin(\lambda/2) & \cos(\lambda/2) \end{pmatrix} \qquad (43)$$

with

$$\lambda/2 = \arccos\left(\sqrt{\frac{\delta+\Omega}{2\Omega}}\right) \; . \tag{44}$$

Hence, one has the identities

$$\begin{aligned}
\Xi^\dagger s_x \Xi &= \frac{\delta}{\Omega}\, s_x + \frac{\sqrt{\Omega^2-\delta^2}}{\Omega}\, s_z \\
\Xi^\dagger s_y \Xi &= s_y \quad , \\
\Xi^\dagger s_z \Xi &= \frac{\delta}{\Omega}\, s_z - \frac{\sqrt{\Omega^2-\delta^2}}{\Omega}\, s_x \; ,
\end{aligned} \tag{45}$$

which may be easily verified.

B. Floquet decomposition for general s

Replacing the spin-$\frac{1}{2}$ operators $s_j = \frac{\sigma_j}{2}$ in the Hamiltonian (34) by their conterparts S_j for general spin quantum number s, one obtains

$$\begin{aligned}
H^{(s)}(t) &= \mathbf{b}(t)\cdot\mathbf{S} \\
&= \omega_0\, S_z + F\,(S_x\,\cos\omega t + S_y\,\sin\omega t) \; .
\end{aligned} \tag{46}$$

According to Sec. II B the general matrix solution to the corresponding Schrödinger equation (33) now is obtained as the lift (32) of the 2×2-matrix (39). Invoking Eqs. (42) and (43), and applying the irrep $R^{(s)}$ to this decomposition yields

$$\begin{aligned}
\Psi^{(s)}(t) &= R^{(s)}\Big(\exp(-\mathrm{i}\omega t\, s_z)\exp(-\mathrm{i}\lambda\, s_y)\exp(-\mathrm{i}\Omega t\, s_z)\Big) \\
&= \exp(-\mathrm{i}\omega t\, S_z)\exp(-\mathrm{i}\lambda\, S_y)\exp(-\mathrm{i}\Omega t\, S_z) \; .
\end{aligned} \tag{47}$$

In order to bring this factorization into the standard Floquet form analogous to Eq. (40),

$$\Psi^{(s)}(t) = P^{(s)}(t)\exp\big(-\mathrm{i}G^{(s)}t\big) \tag{48}$$

with a T-periodic matrix $P^{(s)}(t) = P^{(s)}(t+T)$, we have to distinguish two cases:

(i) For *integer* s we may set

$$\begin{aligned}
P^{(s)}(t) &= \exp(-\mathrm{i}\omega t\, S_z)\,\exp(-\mathrm{i}\lambda\, S_y) \; , \\
G^{(s)} &= \Omega\, S_z \; ,
\end{aligned} \tag{49}$$

obtaining the quasienergies

$$\varepsilon_m = m\,\Omega \quad \mod \omega \tag{50}$$

with integer $m = -s, \ldots, s$.

(ii) For *half-integer* s the requirement that $P^{(s)}(t)$ be T-periodic demands insertion of additional factors $\mathrm{e}^{\pm\mathrm{i}\omega t/2}$, in analogy to the representation (42) for $s = \frac{1}{2}$. This gives

$$\begin{aligned}
P^{(s)}(t) &= \mathrm{e}^{\mathrm{i}\omega t/2}\exp(-\mathrm{i}\omega t\, S_z)\,\exp(-\mathrm{i}\lambda\, S_y) \; , \\
G^{(s)} &= \frac{\omega}{2}\mathbb{1}^{(s)} + \Omega\, S_z \; ,
\end{aligned} \tag{51}$$

where $\mathbb{1}^{(s)}$ denotes the unit matrix in $\mathbb{C}^{2s+1}$. Therefore, the quasienergies now read

$$\varepsilon_m = \frac{\omega}{2} + m\,\Omega \quad \mod \omega \tag{52}$$

for half-integer $m = -s, \ldots, s$.

Denoting the eigenstates of S_z by $|m\rangle$, in both cases *(i)* and *(ii)* the Floquet states can now be written as

$$\begin{aligned}
\Psi^{(s)}(t)\,|m\rangle &= P^{(s)}(t)\,|m\rangle\,\exp(-\mathrm{i}\varepsilon_m t) \\
&\equiv |u_m(t)\rangle\,\exp(-\mathrm{i}\varepsilon_m t) \; ,
\end{aligned} \tag{53}$$

thereby introducing the T-periodic Floquet functions

$$|u_m(t)\rangle = P^{(s)}(t)\,|m\rangle \; . \tag{54}$$

Since $\Omega \to |\omega_0 - \omega|$ when $F \to 0$, the quasienergies (50) and (52) properly connect to the respective eigenvalues of $\omega_0 S_z$ for vanishing driving amplitude. There is, however, an important further distinction to be observed at this point: In either case *(i)* and *(ii)* one finds

$$\varepsilon_m \to \quad m\omega_0 \quad \mod \omega \quad \text{if } \omega < \omega_0 \; , \tag{55}$$

whereas

$$\varepsilon_m \to -m\omega_0 \quad \mod \omega \quad \text{if } \omega > \omega_0 \; . \tag{56}$$

That is, if the driving frequency ω is detuned to the blue side of the transition frequency ω_0, the labeling of the quasienergy representatives (50) and (52) differs from that of the eigenvalues of S_z by a minus sign, effectively reversing their order. This feature needs to be kept in mind for correctly assessing the following results.

We also note that in the adiabatic limit, when the spin is exposed to an arbitrarily slowly varying magnetic field enabling adiabatic following to the instantaneous energy eigenstates, the quasienergies should be given by the one-cyle averages of the instantaneous energy eigenvalues. Indeed, in this limit the Rabi frequency (38) reduces to $\Omega = \sqrt{\omega_0^2 + F^2}$, while the time-independent instantaneous energy levels are $E_m = m\,\Omega$, yielding the expected identity.

IV. THE QUASISTATIONARY DISTRIBUTION

Now we stipulate that the periodically driven spin be coupled to a thermal bath of harmonic oscillators, as sketched in Sec. II A, taking the coupling operator to be of the natural form [39]

$$V = \gamma\, S_x \; . \tag{57}$$

In order to calculate the Fourier decompositions (7) of the Floquet matrix elements of V, and referring to the above representation (54) of the Floquet functions, we thus need to consider the operator

$$\begin{aligned}
& P^{(s)\dagger}(t)\, S_x\, P^{(s)}(t) \\
=\; & \exp(\mathrm{i}\lambda\, S_y)\exp(\mathrm{i}\omega t\, S_z)\; S_x\; \exp(-\mathrm{i}\omega t\, S_z)\exp(-\mathrm{i}\lambda\, S_y) \\
\equiv\; & \sum_{\ell\in\mathbb{Z}} V^{(\ell)}\exp(\mathrm{i}\ell\omega t) \; ;
\end{aligned} \tag{58}$$

note that the additional phase factor $\mathrm{e}^{\mathrm{i}\omega t/2}$ contained in the expression (51) for $P^{(s)}(t)$ with half-integer s cancels here. Using the $su(2)$ commutation relations and their counterparts for general s, we deduce

$$\begin{aligned} & \exp\left(\mathrm{i}\omega t\, S_z\right)\; S_x\; \exp\left(-\mathrm{i}\omega t\, S_z\right) \\ = \; & \frac{1}{2}\left(\mathrm{e}^{\mathrm{i}\omega t}\, S_+ + \mathrm{e}^{-\mathrm{i}\omega t}\, S_-\right) \; . \end{aligned} \tag{59}$$

Hence, as in the case $s = \frac{1}{2}$ studied in Ref. [29], the only non-vanishing Fourier components $V^{(\ell)}$ occur for $\ell = \pm 1$:

$$V^{(\pm 1)} = \frac{\gamma}{2} \exp\left(\mathrm{i}\lambda\, S_y\right) S_\pm \exp\left(-\mathrm{i}\lambda\, S_y\right) \; . \tag{60}$$

Applying $R^{(s)}$ to Eqs. (45), this yields

$$V^{(\pm 1)} = \frac{\gamma}{2}\left(\frac{\delta}{\Omega}\, S_x \, + \frac{\sqrt{\Omega^2 - \delta^2}}{\Omega}\, S_z \, \pm \mathrm{i}\, S_y\right) \; . \tag{61}$$

Thus, $V^{(\pm 1)}$ is a tridiagonal matrix. For computing the partial transition rates (19) we therefore have to consider only frequencies $\omega_{mn}^{(\pm 1)}$ of pseudotransitions [29], for which $m = n$, and of transitions between neighboring Floquet states, $m = n \pm 1$. In view of the definition (9), and taking the quasienergies given by Eqs. (50) and (52) without "mod ω" as the canonical representatives of their respective quasienergy classes (5), one has

$$\omega_{mn}^{(\pm 1)} = \left\{ \begin{array}{rcl} \pm\omega & : & m = n \; , \\ \Omega \pm \omega & : & m = n + 1 \; , \\ -\Omega \pm \omega & : & m = n - 1 \; . \end{array} \right. \tag{62}$$

According to the Pauli master equation (20), the quasistationary distribution $\{p_m\}_{m=s,\ldots,-s}$ which establishes itself under the combined influence of time-periodic driving and the thermal oscillator bath is the eigenvector of a tridiagonal matrix $\widetilde{\Gamma}$ corresponding to the eigenvalue 0, where $\widetilde{\Gamma}$ is obtained from $\Gamma \equiv \Gamma^{(1)} + \Gamma^{(-1)}$ by subtracting from the diagonal elements the respective column sums, *i.e.*,

$$\widetilde{\Gamma}_{mn} = \Gamma_{mn} - \delta_{mn} \sum_{k=-s}^{s} \Gamma_{kn} \; . \tag{63}$$

Since $\widetilde{\Gamma}$ is tridiagonal with non-vanishing secondary diagonal elements, this eigenvector is unique up to normalization. Moreover, it is evident that we only need the matrix elements of Γ in the secondary diagonals for calculating the quasistationary distribution, whereas the diagonal elements will be required for computing the dissipation rate [29].

The very fact that $V^{(\pm 1)}$, and hence Γ, merely is a tridiagonal matrix has a conceptually important consequence: It enforces detailed balance, meaning that each term of the sum (20) vanishes individually. With Γ being tridiagonal, this sum reduces to

$$\begin{aligned} & \left(\Gamma_{n,n-1}\, p_{n-1} - \Gamma_{n-1,n}\, p_n\right) \\ + & \left(\Gamma_{n,n+1}\, p_{n+1} - \Gamma_{n+1,n}\, p_n\right) \; = \; 0 \end{aligned} \tag{64}$$

for all $n = -s+1, \ldots, s-1$, since the term with $m = n$ in Eq. (20) cancels. In the border cases $n = -s$ or $n = s$ this identity still holds, but only one bracket survives. Upon setting the first bracket in this Eq. (64) to zero, one obtains

$$\frac{p_n}{p_{n-1}} = \frac{\Gamma_{n,n-1}}{\Gamma_{n-1,n}} \tag{65}$$

for $n = -s+1, \ldots, s-1$. Together with the normalization requirement, this relation already determines the entire distribution $\{p_m\}$. In particular, it entails

$$\frac{p_{n+1}}{p_n} = \frac{\Gamma_{n+1,n}}{\Gamma_{n,n+1}} \; , \tag{66}$$

thus ensuring that also the second bracket in Eq. (64) vanishes, confirming detailed balance.

A further factor of substantial importance is the spectral density $J(\widetilde{\omega})$, which may allow one to manipulate the quasistationary distribution to a considerable extent. For the sake of simplicity and transparent discussion, here we assume that $J(\widetilde{\omega}) \equiv J_0$ is constant. The distinction between the physically different positive and negative transition frequencies, which lead to the two different expressions (15) and (17) entering the transition rates (19), now prompts us to treat the cases $0 < \omega < \Omega$ and $0 < \Omega < \omega$ separately, while the resonant case $\omega = \Omega$ can be dealt with by means of limit procedures.

A. Low-frequency case $0 < \omega < \Omega$

In order to utilize the above Eq. (66) for determining the distribution $\{p_m\}$ recursively we only need to evaluate the partial rates $\Gamma_{m,m+1}^{(\pm 1)}$ and $\Gamma_{m+1,m}^{(\pm 1)}$ according to the general prescription (19). In the low-frequency case $0 < \omega < \Omega$ this leads to the expressions

$$\begin{aligned} \Gamma_{m,m+1}^{(\pm 1)} \; &= \; \frac{s(s+1) - m(m+1)}{16\left(1 - \mathrm{e}^{-\beta(\Omega \mp \omega)}\right)} \left(\frac{\Omega \mp \delta}{\Omega}\right)^2 \\ \Gamma_{m+1,m}^{(\pm 1)} \; &= \; \frac{s(s+1) - m(m+1)}{16\left(\mathrm{e}^{\beta(\Omega \pm \omega)} - 1\right)} \left(\frac{\Omega \pm \delta}{\Omega}\right)^2 \end{aligned} \tag{67}$$

which have been scaled by $\Gamma_0 = 2\pi\gamma^2 J_0$, and have thus been made dimensionless. Evidently, these representations imply that the desired ratio

$$\frac{\Gamma_{m+1,m}}{\Gamma_{m,m+1}} \equiv q_L \tag{68}$$

is independent of both s and m; a tedious but straightforward calculation readily yields

$$q_L = \frac{2\delta\Omega\sinh(\beta\omega) + \left(\delta^2 + \Omega^2\right)\left(\mathrm{e}^{-\beta\Omega} - \cosh(\beta\omega)\right)}{2\delta\Omega\sinh(\beta\omega) - \left(\delta^2 + \Omega^2\right)\left(\mathrm{e}^{\beta\Omega} - \cosh(\beta\omega)\right)} \; . \tag{69}$$

Therefore, not only does one find detailed balance here, but the occupation probabilities even generate a finite geometric sequence: By construction, the geometric sequence

$$\mathbf{q}_L \equiv (q_L^{2s}, q_L^{2s-1}, \ldots, q_L, 1) \tag{70}$$

is an eigenvector of the matrix $\widetilde{\Gamma}$ introduced in Eq. (63) with eigenvalue 0, and hence yields the desired quasistationary Floquet-state occupation probabilites $\{p_m\}$ after normalization.

B. High-frequency case $0 < \Omega < \omega$

Analogously, in the high-frequency case $0 < \Omega < \omega$ we require the dimensionless partial rates

$$\begin{aligned} \Gamma^{(\pm 1)}_{m,m+1} &= \pm \frac{s(s+1) - m(m+1)}{16\left(\mathrm{e}^{\beta(\pm\omega-\Omega)} - 1\right)} \left(\frac{\Omega \mp \delta}{\Omega}\right)^2 \\ \Gamma^{(\pm 1)}_{m+1,m} &= \pm \frac{s(s+1) - m(m+1)}{16\left(\mathrm{e}^{\beta(\Omega\pm\omega)} - 1\right)} \left(\frac{\Omega \pm \delta}{\Omega}\right)^2 \end{aligned} \tag{71}$$

for constructing the matrix $\Gamma = \Gamma^{(1)} + \Gamma^{(-1)}$. Once more, the ratio

$$\frac{\Gamma_{m+1,m}}{\Gamma_{m,m+1}} \equiv q_H \tag{72}$$

does depend neither on s nor on m, so that the occupation probabilities again form a geometric sequence; after some juggling, one finds

$$q_H = \frac{\left(\delta^2 + \Omega^2\right)\sinh(\beta\omega) + 2\delta\Omega\left(\mathrm{e}^{-\beta\Omega} - \cosh(\beta\omega)\right)}{\left(\delta^2 + \Omega^2\right)\sinh(\beta\omega) - 2\delta\Omega\left(\mathrm{e}^{\beta\Omega} - \cosh(\beta\omega)\right)} \, . \tag{73}$$

This quantity determines the solution vector

$$\mathbf{q}_H \equiv (q_H^{2s}, q_H^{2s-1}, \ldots, q_H, 1) \tag{74}$$

to the equation $\widetilde{\Gamma}\,\mathbf{q}_H = \mathbf{0}$ in the high-frequency regime, providing the corresponding quasistationary Floquet distribution upon normalization.

C. The quasitemperature

For ease of notation, let us define

$$q \equiv \begin{cases} q_L & : \quad 0 < \omega < \Omega \\ q_H & : \quad 0 < \Omega < \omega \, . \end{cases} \tag{75}$$

Recalling $\Omega \to |\omega_0 - \omega|$ in the limit $F \to 0$ of vanishing driving amplitude, one then finds

$$q \to \begin{cases} \mathrm{e}^{-\beta\omega_0} & : \quad \omega < \omega_0 \\ \mathrm{e}^{+\beta\omega_0} & : \quad \omega > \omega_0 \, . \end{cases} \tag{76}$$

Hence, in the absence of the time-periodic driving field the above solutions (70) and (74) correctly lead to a Boltzmann distribution with the inverse bath temperature β, thereby indicating thermal equilibrium of the spin system; the unfamiliar "plus"-sign appearing in this limit (76) in the case of blue detuning merely reflects the reversed labeling (56).

More importantly, in both cases $0 < \omega < \Omega$ and $0 < \Omega < \omega$ the occupation probabilities $\{p_m\}$ of the Floquet states constitute a geometric sequence even under arbitrarily strong driving, when the system is far remote from usual thermal equilibrium. Since the quasienergies (50) and (52) are equidistant, the quasistationary distribution can therefore still be regarded as a Boltzmann distribution, but now being parametrized by an effective quasitemperature:

$$p_m = \frac{1}{Z_1} q^{s+m} = \frac{1}{Z_2} \mathrm{e}^{-\vartheta\varepsilon_m} = \frac{1}{Z_q} \mathrm{e}^{-\vartheta\Omega m} \tag{77}$$

with $m = s, \ldots, -s$ for both integer and half-integer s, where the factors Z_1, Z_2, and Z_q are adjusted to ensure the normalization $\sum_{m=-s}^{s} p_m = 1$, and

$$\vartheta \equiv -\frac{\ln q}{\Omega} \tag{78}$$

is the inverse quasitemperature of the periodically driven spin system coupled to an oscillator bath with inverse regular temperature β. When defining the quasitemperature in this analytically convenient manner (78) we do not distinguish between the cases (76) of red and blue detuning, so that our quasitemperature will be negative under weak driving with blue detuning. In accordance with the distinction (75) we will also designate the inverse quasitemperature as ϑ_L and ϑ_H, respectively. Note that the parametrization of the geometric distribution (77) in terms of the system's quasienergies and a quasitemperature, in formal analogy to the canonical distribution of equilibrium thermodynamics [28], requires that the "modulo ω-indeterminacy" of the quasienergies has been resolved in some way, so that the quasienergies entering this distribution refer to particular, well defined representatives of their classes (5); here we have selected the representatives given by Eqs. (50) and (52) without "mod ω". This implies that the definition of the quasitemperature still contains a certain degree of arbitrariness. Notwithstanding this remark, the very distribution $\{p_m\}$ itself, governing the observable physics, is defined uniquely.

The dimensionless inverse quasitemperature $\omega_0\vartheta$ ultimately depends on the scaled driving amplitude F/ω_0, the scaled driving frequency ω/ω_0, and the dimensionless inverse actual temperature $\omega_0\beta$ of the heat bath, but not on the spin quantum number s. In contrast, the partition function Z_q depends on s:

$$Z_q = \sum_{m=-s}^{s} \exp(-\vartheta\Omega m) = \frac{\sinh\left(\dfrac{2s+1}{2}\,\vartheta\Omega\right)}{\sinh\left(\dfrac{\vartheta\Omega}{2}\right)} \, . \tag{79}$$

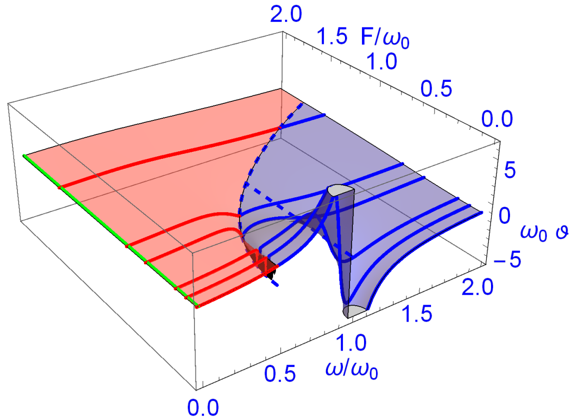

FIG. 1: Dimensionless inverse quasitemperature $\omega_0\vartheta$ of the spin system under the influence of a circularly polarized monochromatic driving force with amplitude F and frequency ω, being coupled to a harmonic-oscillator bath of inverse actual temperature $\omega_0\beta = 1$. The blue part of the graph corresponds to the high-frequency regime $0 < \Omega < \omega$, the red part to the low-frequency regime $0 < \omega < \Omega$. Along the line segment $\omega = \omega_0$ with $F < \omega_0$ and along the parabola $\omega = \omega_c$ given by Eq. (80), both marked by blue dashes, the inverse quasitemperature vanishes. A few functions $\vartheta(\omega)$ for constant F are highlighted, showing a kink at $\omega = \omega_c$ according to Eqs. (82). The limit $\omega_0\vartheta = \omega_0\beta$ for $\omega/\omega_0 \to 0$ is indicated by the green line.

The inverse quasitemperature ϑ vanishes — meaning that the periodically driven system effectively becomes infinitely hot, so that all its Floquet states are populated equally — regardless of the bath temperature, if either $\omega = \omega_0$ while $0 < F < \omega_0$, or if

$$\omega = \omega_c \equiv \frac{1}{2\omega_0}\left(F^2 + \omega_0^2\right) \ , \tag{80}$$

where the latter equation defines the boundary $\omega = \Omega$ between the low- and the high-frequency regime, see Fig. 1. At this boundary there is a continuous change from ϑ_L to ϑ_H, since

$$\lim_{\omega\uparrow\omega_c} \vartheta_L = \lim_{\omega\downarrow\omega_c} \vartheta_H = 0 \, . \tag{81}$$

However, the two functions ϑ_L and ϑ_H do not join smoothly at $\omega = \omega_c$, since their derivatives with respect to ω adopt limits with opposite signs:

$$\begin{aligned} \lim_{\omega\uparrow\omega_c} \frac{\partial\vartheta_L}{\partial\omega} &= -\frac{4\beta\omega_0^3\left(F^4+\omega_0^4\right)}{F^4\left(F^2+\omega_0^2\right)^2} \, , \\ \lim_{\omega\downarrow\omega_c} \frac{\partial\vartheta_H}{\partial\omega} &= \frac{4\beta\omega_0^3\left(F^4+\omega_0^4\right)}{F^4\left(F^2+\omega_0^2\right)^2} \, . \end{aligned} \tag{82}$$

We will now investigate the behavior of the function $\omega_0\vartheta(\omega_0\beta, \omega/\omega_0, F/\omega_0)$ in the limits corresponding to the four sideways faces of the box bounding the plot displayed in Fig. 1:

(i) As already noted at the end of Sec. III B, in the low-frequency limit $\omega/\omega_0 \to 0$ the canonical representatives of the quasienergies (50) and (52) approach the actual energies $E_m = m\sqrt{\omega_0^2 + F^2}$ (with $m = s, \dots, -s$) of a spin exposed to a slowly varying drive. Hence, in this limit the "periodic thermodynamics" investigated here must reduce to the usual thermodynamics described by a canonical ensemble; in particular, the inverse quasitemperature ϑ must approach the true inverse bath temperature β. This expectation is borne out by the leading term of the low-frequency expansion

$$\begin{aligned} \omega_0\vartheta_L &= \omega_0\beta + \frac{2\omega_0\beta(\omega/\omega_0)}{(F/\omega_0)^2+2} \\ &\quad + \frac{\omega_0\beta(\omega/\omega_0)^2}{2\big((F/\omega_0)^2+2\big)^2}\left(8 - 4(F/\omega_0)^2 - \frac{(F/\omega_0)^4\,\omega_0\beta\,\coth\left(\frac{\omega_0\beta}{2}\sqrt{(F/\omega_0)^2+1}\right)}{\sqrt{(F/\omega_0)^2+1}}\right) + O(\omega/\omega_0)^3 \, . \end{aligned} \tag{83}$$

(ii) Next we consider the ultrahigh-frequency limit $\omega/\omega_0 \to \infty$, keeping both $\omega_0\beta$ and F/ω_0 fixed. Inspecting q_H as defined by Eq. (73), and observing that asymptotically $\Omega = \sqrt{(\omega_0-\omega)^2 + F^2} \sim \omega$ for $\omega/\omega_0 \to \infty$, one finds $q_H \to 1$, and hence

$$\lim_{\omega/\omega_0\to\infty} \omega_0\vartheta_H \;=\; 0 \; ; \tag{84}$$

as shown by Fig. 1, this limit is approached with negative quasitemperatures. This means that for high driving frequencies all Floquet states are populated almost equally, independent of both the driving amplitude and the bath temperature.

(iii) The static limit of vanishing driving amplitude, $F/\omega_0 \to 0$, yields

$$\lim_{F/\omega_0\to 0} \omega_0\vartheta_L \;=\; \lim_{F/\omega_0\to 0} \omega_0\vartheta_H \;=\; \frac{\omega_0\beta}{1-\omega/\omega_0} \, . \tag{85}$$

Once again, this apparently strange expression, exhibiting a pole at $\omega = \omega_0$ which is prominently visible in Fig. 1,

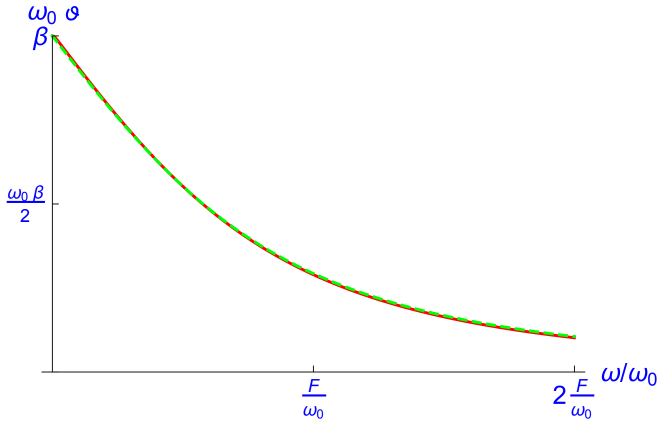

FIG. 2: Inverse dimensionless quasitemperature $\omega_0\vartheta$ for $\omega_0\beta = 1$ and $F/\omega_0 = 100$ as a function of ω/ω_0 in the regime $0 < \omega/\omega_0 < 2F/\omega_0$. We show the exact values of $\omega_0\vartheta_L$ (red line), as well as the asymptotic form (86) (green dashes).

is fully in agreement with the expectation that the "periodic thermodynamics" should reduce to ordinary thermodynamics when the time-periodic driving force vanishes. Namely, for $F/\omega_0 \to 0$ the system possesses the energy eigenvalues $E_m = m\omega_0$, whereas the quasienergies (50) or (52) reduce to $\varepsilon_m = m|\omega_0 - \omega|$ or $\varepsilon_m = \omega/2 + m|\omega_0 - \omega|$ with $m = s, \ldots, -s$, while the Floquet states approach the energy eigenstates. Since the parametrization of their occupation probabilities in terms of either the distribution (77) or the standard canonical distribution then must lead to identical values, one obtains the requirement $\beta\omega_0 = \vartheta(\omega_0 - \omega)$ for $0 < \omega < \omega_0$, which immediatly furnishes the above limit. In the case of blue detuning, for $0 < \omega_0 < \omega$, the formal inversion of the quasienergy levels expressed by Eq. (56) yields an an additional "minus" sign, again leading to the limit (85).

(iv) In the converse strong-driving limit $F/\omega_0 \to \infty$ we first focus on the regime $\omega \sim F$ where $\vartheta = \vartheta_L > 0$. After some transformations we obtain the asymptotic form

$$\omega_0\vartheta_L \sim \omega_0\beta - \frac{\omega\beta}{\sqrt{(F/\omega_0)^2 + (\omega/\omega_0)^2}} \tag{86}$$

in this regime, so that the inverse quasitemperature decreases monotonically with increasing driving amplitude, as exemplified by Fig. 2.

In contrast, for $\omega > \omega_c$ as given by Eq. (80) we have $\vartheta = \vartheta_H < 0$. Asymptotically, here we find

$$\omega_0\vartheta_H \sim -\frac{\omega_0\beta}{\omega/\omega_0} + \frac{\omega_0\beta}{2(\omega/\omega_0)^2}\,(F/\omega_0)^2 \; . \tag{87}$$

This asymptotic function exhibits a pronounced minimum at $\omega/\omega_0 = (F/\omega_0)^2$ of depth $\omega_0\vartheta_H \sim -\omega_0^3\beta/(2F^2)$, depicted in Fig. 3.

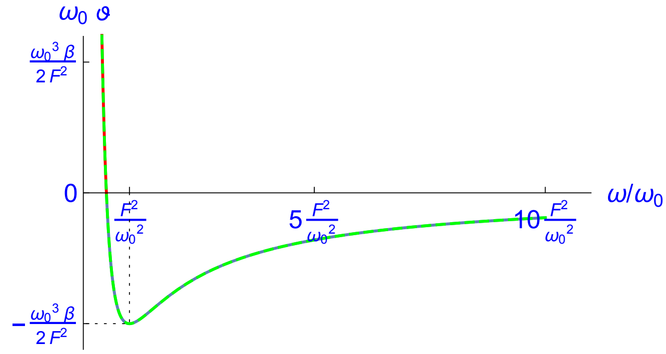

FIG. 3: Inverse dimensionless quasitemperature $\omega_0\vartheta$ for $\omega_0\beta = 1$ and $F/\omega_0 = 100$ as a function of ω/ω_0 in the regime $\omega/\omega_0 > (F/\omega_0)^2/2$. We show the exact values of $\omega_0\vartheta_L$ (red line) and $\omega_0\vartheta_H$ (blue line), together with the asymptotic form (87) (green dashes).

V. APPLICATION: PERIODICALLY DRIVEN PARAMAGNETS

A. Calculation of quasithermal expectation values

Starting from the proposition that the Floquet states of a periodically driven spin system be populated according to the distribution (77) we introduce the quasithermal average of the spin component in the direction of the static field,

$$\langle S_z\rangle_q \equiv \frac{1}{Z_2}\sum_{m=-s}^{s}\langle u_m(t)|S_z|u_m(t)\rangle\,\exp(-\vartheta\varepsilon_m) \; . \tag{88}$$

In order to evaluate this expression we utilize the representation (54) of the Floquet functions, giving

$$\begin{aligned} Z_2\langle S_z\rangle_q &= \sum_{m=-s}^{s}\langle m|P^{(s)\dagger}\,S_z\,P^{(s)}|m\rangle\,\exp(-\vartheta\varepsilon_m) \\ &= \mathrm{Tr}\left(P^{(s)\dagger}\,S_z\,P^{(s)}\,\exp\bigl(-\vartheta G^{(s)}\bigr)\right) \; . \end{aligned} \tag{89}$$

Next, we resort once more to the lifting technique: For $s = \frac{1}{2}$, the decomposition (42) readily yields

$$\begin{aligned} P^\dagger s_z P &= \Xi^\dagger s_z \Xi \\ &\overset{(45)}{=} \frac{\delta}{\Omega}\,s_z - \frac{\sqrt{\Omega^2-\delta^2}}{\Omega}\,s_x \; . \end{aligned} \tag{90}$$

Applying the irrep $r^{(s)}$, we deduce

$$\begin{aligned} P^{(s)\dagger}\,S_z\,P^{(s)} &= r^{(s)}\bigl(P^\dagger s_z P\bigr) \\ &= \frac{\delta}{\Omega}\,S_z - \frac{\sqrt{\Omega^2-\delta^2}}{\Omega}\,S_x \; . \end{aligned} \tag{91}$$

Inserting this into the above identity (89), and calculating the trace in the eigenbasis of S_z, we obtain the important result

$$\langle S_z\rangle_q = \frac{1}{Z_q}\frac{\omega_0-\omega}{\Omega}\sum_{m=-s}^{m} m\,\mathrm{e}^{-\vartheta\Omega\,m} \; , \tag{92}$$

valid for both integer and half-integer s. Although the unusual-looking prefactor $(\omega_0 - \omega)/\Omega$ indeed implies that the z-component of the magnetization vanishes for $\omega = \omega_0$, it does not imply that the magnetization reverses its direction when ω is varied across ω_0, since the reversal of the prefector's sign can be compensated by a simultaneous change of the sign of the quasitemperature, as it happens for low driving amplitudes according to Eq. (85).

B. Response of paramagnetic materials to circularly polarized driving fields

As an experimentally accessible example of the above considerations, and thus as a possible laboratory application of periodic thermodynamics, we consider the magnetization of an ideal paramagnetic substance under the influence of both a static magnetic field applied in the z-direction, and a circularly polarized oscillating magnetic field applied in the x-y-plane. In order to facilitate comparison with the literature, here we re-install the Planck constant $\hbar$ and the Boltzmann constant k_B.

We assume that the magnetic atoms of the substance have an electron shell with total angular momentum J, resulting from the coupling of orbital angular momentum and spin, giving the magnetic moment $\mu = -g_J \mu_\mathrm{B} J$. Here μ_B denotes the Bohr magneton, and $g_J > 0$ is the Landé g-factor. In the presence of a constant magnetic field B_0 this moment gives rise to the energy levels

$$E_m = m\, g_J \mu_\mathrm{B} B_0 \equiv m\, \hbar\omega_0 \; , \tag{93}$$

where $m = -J \ldots, J$ is the magnetic quantum number, with the "plus"-sign accounting for the fact that the magnetic moment tends to align parallel to the applied magnetic field, favoring $m = -J$.

Let us briefly recall the usual textbook treatment of the ensuing thermal paramagnetism within the canonical ensemble [27, 28], assuming the substance to possess a temperature T. Then the canonical partition function

$$\begin{aligned} Z_0 &= \sum_{m=-J}^{J} \exp\left(-\frac{E_m}{k_\mathrm{B} T}\right) = \sum_{m=-J}^{J} \exp\left(-m\frac{\hbar\omega_0}{k_B T}\right) \\ &= \frac{\sinh\left(\dfrac{2J+1}{2J}\, y_0\right)}{\sinh\left(\dfrac{y_0}{2J}\right)} \end{aligned} \tag{94}$$

which depends on the dimensionless quantity

$$y_0 \equiv \frac{g_J \mu_\mathrm{B} B_0}{k_\mathrm{B} T}\, J = \frac{\hbar\omega_0}{k_\mathrm{B} T}\, J \tag{95}$$

serves as moment-generating function, in the sense that the thermal expectation value of the magnetization M is obtained by taking the appropriate derivative of its logarithm, namely,

$$\begin{aligned} \langle M \rangle &= \frac{N}{V}\, \langle \mu \rangle = -\frac{N}{V}\, g_J \mu_\mathrm{B} \langle m \rangle \\ &= \frac{N}{V} \frac{\partial}{\partial B_0}\, k_B T \ln Z_0 \; . \end{aligned} \tag{96}$$

Here $\frac{N}{V}$ denotes the density of contributing atoms. Working out this prescription, one finds the magnetization [27, 28]

$$\langle M \rangle = M_0\, B_J(y_0) \; , \tag{97}$$

where

$$M_0 = \frac{N}{V}\, g_J \mu_\mathrm{B} J \tag{98}$$

denotes the saturation magnetization, and

$$B_J(y) \equiv \frac{2J+1}{2J} \coth\left(\frac{2J+1}{2J} y\right) - \frac{1}{2J} \coth\left(\frac{y}{2J}\right) \tag{99}$$

is the so-called Brillouin function of order J [25]; this theoretical prediction (97) has been beautifully confirmed in low-temperature experiments with paramagnetic ions by Henry [26] already in 1952. In the weak-field limit $\mu_\mathrm{B} B_0 \ll k_\mathrm{B} T$ one may use to approximation

$$B_J(y) \approx \frac{J+1}{J} \frac{y}{3} \quad \text{for} \quad 0 < y \ll 1 \; , \tag{100}$$

giving

$$\langle M \rangle \approx \frac{N}{V} \frac{(g_J \mu_\mathrm{B})^2 J(J+1)}{3 k_\mathrm{B} T}\, B_0 \; . \tag{101}$$

Returning to periodic thermodynamics, let us add the circularly polarized field $B_1(\cos\omega t, \sin\omega t, 0)$ perpendicular to the constant one. Then the Rabi frequency (38) can be written as

$$\Omega = \sqrt{(\omega_0 - \omega)^2 + (g_J \mu_\mathrm{B} B_1/\hbar)^2} \; , \tag{102}$$

where

$$\omega_0 = \frac{g_J \mu_\mathrm{B} B_0}{\hbar} \tag{103}$$

measures the strength of the static field in accordance with Eq. (93). Also note that the sign of the time-independent contribution $\omega_0 S_z$ to the Hamiltonian (46) now has to be inverted, in order to account for the proper ordering of the energy eigenvalues (93). Assuming that the spins' environment is correctly described by a thermal oscillator bath with constant spectral density J_0, so that the distribution (77) governs the Floquet-state occupation probabilities, we can invoke the above result (92) to write the observable magnetization in the form

$$\begin{aligned} \langle M \rangle_q &= -\frac{N}{V}\, g_J \mu_\mathrm{B} \langle S_z \rangle_q \\ &= -\frac{N}{V} \frac{g_J \mu_\mathrm{B}}{Z_q} \frac{\omega_0 - \omega}{\Omega} \sum_{m=-J}^{J} m \, \exp\left(-m\frac{\hbar\Omega}{k_\mathrm{B}\tau}\right) \; , \end{aligned} \tag{104}$$

where $\tau = 1/(k_B\vartheta)$ is the quasitemperature, and

$$Z_q = \sum_{m=-J}^{J} \exp\left(-m\frac{\hbar\Omega}{k_B\tau}\right) \qquad (105)$$

is the corresponding partition function (79). Quite remarkably, this expression (104) is a perfect formal analog of the previous Eq. (96), since we have

$$\langle M\rangle_q = \frac{N}{V}\frac{\partial}{\partial B_0}k_B\tau\ln Z_q\ , \qquad (106)$$

taking into account the nonlinear dependence of the Rabi frequency (102) on the static field strength B_0. Hence, the resulting quasithermal magnetization can be expressed in a manner analogous to Eq. (97), namely,

$$\langle M\rangle_q = M_1\,B_J(y_1)\ , \qquad (107)$$

with modified saturation magnetization

$$M_1 = \frac{\omega_0-\omega}{\Omega}M_0\ , \qquad (108)$$

and the argument of the Brillouin function now depending on the quasitemperature,

$$y_1 = \frac{\hbar\Omega}{k_B\tau}J\ . \qquad (109)$$

For consistency, this prediction (107) must reduce to the usual weak-field magnetiziation (101) when both $B_1 \to 0$ and $\mu_B B_0 \ll k_B T$. This is ensured by the limit (85): For sufficiently small B_1, the quasitemperature τ is related to the actual bath temperature T through

$$\frac{1}{k_B\tau} \approx \frac{\omega_0}{\omega_0-\omega}\frac{1}{k_B T}\ . \qquad (110)$$

Inserting this into Eq. (107), one can employ the approximation (100) for frequencies ω not too close to ω_0. In this way one recovers the expected expression (101) unless $\omega \approx \omega_0$, in which case one has $\langle M\rangle_q \approx 0$.

As is evident from the above discussion, under typical ESR conditions with weak driving amplitudes, such that B_1/B_0 is on the order of 10^{-2} or less [40], the difference between the quasithermal magnetization $\langle M\rangle_q$ and the customary thermal magnetization $\langle M\rangle$ is more or less negligible, except for driving frequencies close to resonance. This is illustrated in Fig. 4 for $g_J\mu_B B_1/(\hbar\omega) = 0.01$, where we have chosen the bath temperature according to the fixed driving frequency, $k_B T = \hbar\omega$. The vanishing of the magnetization at $\omega_0 = \omega$ is due to the prefactor $\omega_0 - \omega$ in (108). The second sharp dip at $\omega_0 \approx 2\omega$ can be explained by the vanishing of the inverse quasitemperature ϑ, see (80). Other measurable effects have to be expected in the strong-driving regime.

A particularly striking observation can be made in Fig. 5, where $k_B T = \hbar\omega_0$ and $J = 1$: Under strong driving, there the ratio $\langle M\rangle_q/\langle M\rangle$ actually becomes *negative*

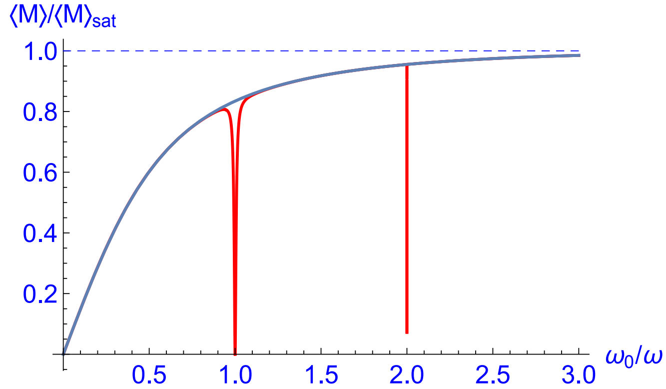

FIG. 4: Magnetization $\langle M\rangle$ divided by the saturation magnetization (98) as a function of ω_0/ω. The blue curve represents the ordinary thermal magnetization (97), the red one the quasithermal magnetization (107) for weak driving, $g_J\mu_B B_1/(\hbar\omega) = 0.01$. Here we have set $k_B T = \hbar\omega$ and $J = 7/2$.

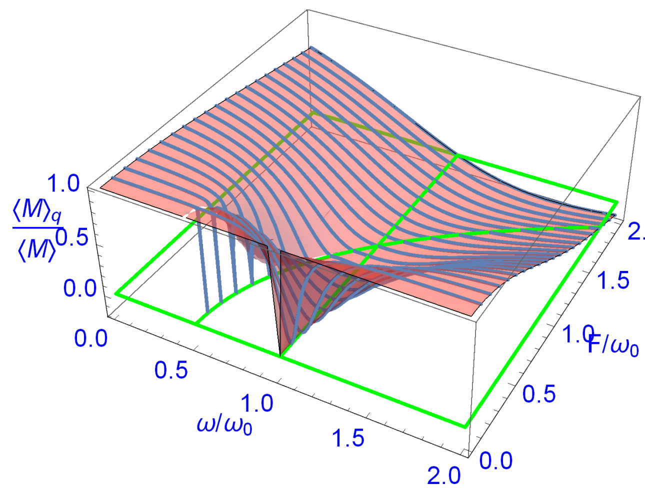

FIG. 5: Ratio $\langle M\rangle_q/\langle M\rangle$ of the quasithermal magnetization (107) and the customary magnetization (97) as function of ω/ω_0 and F/ω_0, with $F = g_J\mu_B B_1/\hbar$. Parameters chosen here are $k_B T = \hbar\omega_0$ and $J = 1$. Along the green line $\omega = \omega_0$ and along the green parabola $\omega = \omega_c$ given by Eq. (80) the quasithermal magnetization vanishes, so that $\langle M\rangle_q/\langle M\rangle$ becomes negative for strong driving with frequencies $\omega_0 < \omega < \omega_c$.

for frequencies $\omega_0 < \omega < \omega_c$, implying that the paramagnetic material effectively becomes a diamagnetic one. The possibility of turning a paramagnet into a diamagnet through the application of strong time-periodic forcing is a "hard" prediction of periodic thermodynamics which now awaits its experimental verification.

VI. DISSIPATION

Since a bath-induced transition from a Floquet state n to a Floquet state m is accompanied by all frequencies

$\omega_{mn}^{(\ell)}$ as introduced in Eq. (9), the rate of energy dissipated in the quasistationary state is given by [29]

$$R = -\sum_{mn\ell} \hbar\omega_{mn}^{(\ell)}\,\Gamma_{mn}^{(\ell)}\,p_n \; . \tag{111}$$

This expression can now be evaluated for all spin quantum numbers s. In addition to the partial transition rates (19) for neigboring Floquet states $m = n \pm 1$ listed in Secs. IV A and IV B, Eq. (111) also requires the rates for pseudotransitions with $m = n$. Again dividing by $\Gamma_0 = 2\pi\gamma^2 J_0$, we obtain the dimensionless diagonal transition rates

$$\Gamma_{mm}^{(\pm 1)} = \pm\frac{(2m)^2 F^2}{16\left(\mathrm{e}^{\pm\beta\omega}-1\right)\Omega^2} \tag{112}$$

for $m = s, \ldots, -s$, valid for both cases $0 < \omega < \Omega$ and $0 < \Omega < \omega$. In order to represent R in a condensed fashion we define the polynomials

$$\begin{aligned} P_s(q) &\equiv -2\sum_{m=0}^{2s}(m-s)^2 q^m \\ Q_s(q) &\equiv \frac{1}{2}\sum_{m=0}^{2s-1}(m+1)(2s-m)q^m \\ z_s(q) &\equiv \sum_{m=0}^{2s} q^m \; , \end{aligned} \tag{113}$$

together with the expression

$$A^{\pm}(q) \equiv \frac{\left(\mathrm{e}^{\beta(\pm\omega+\Omega)}q - 1\right)(\delta\pm\Omega)^2(\omega\pm\Omega)}{\mathrm{e}^{\beta(\pm\omega+\Omega)}-1} \; . \tag{114}$$

Dividing by $\omega_0\Gamma_0$, one obtains a dimensionless dissipation rate which can now be written in the form

$$R = \frac{P_s(q)\,\omega\,F^2 + Q_s(q)\left(A^+(q) \mp A^-(q)\right)}{8\,z_s(q)\,\Omega^2} \; , \tag{115}$$

where one has to insert either q_L or q_H for q, in accordance with the case distinction (75), and the "$\mp$"-sign in the numerator becomes "minus" for $0 < \omega < \Omega$, but "plus" for $0 < \Omega < \omega$.

After resolving all symbols R will be a function of five arguments, $R = R(s, \beta, \omega, \omega_0, F)$, which makes the discussion more difficult than in the case of $s = \frac{1}{2}$ that has been considered in Ref. [29]. Thus, here we mention only the most perspicuous aspects of the dissipation function. Generally, we observe that the dissipation rate is always non-negative, $R \geq 0$, but a proof of this is beyond the scope of the present article and will be published elsewhere.

Recall that for both $\omega = \omega_0$ with $0 < F < \omega_0$ and $\omega = \omega_c = \left(F^2 + \omega_0^2\right)/(2\omega_0)$ we have $q = 1$, and hence $\vartheta = 0$. It turns out that along these two curves in the $(\omega,\, F)$–plane the dimensionless rate R takes on the value

$$R_0 = \frac{1}{6}s(s+1) \; , \tag{116}$$

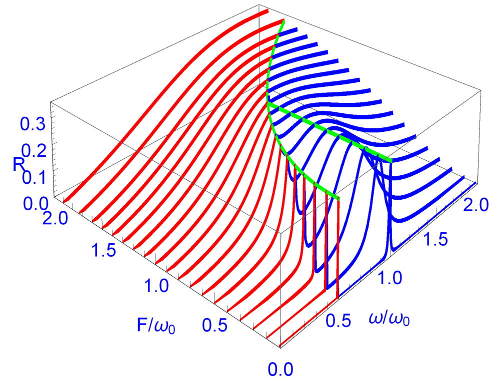

FIG. 6: The dimensionless dissipation rate R for $s = 1$ and $\omega_0\beta = 1$ as a function of ω/ω_0 and F/ω_0. The blue part of the graph corresponds to the high-frequency regime $0 < \Omega < \omega$, the red one to the low-frequency regime $0 < \omega < \Omega$. Along the line $\omega = \omega_0$ with $0 < F < \omega_0$ and along the parabola $\omega = \omega_c$ given by Eq. (80) the dissipation rate takes on the constant value $s(s+1)/6 = 1/3$ (green lines).

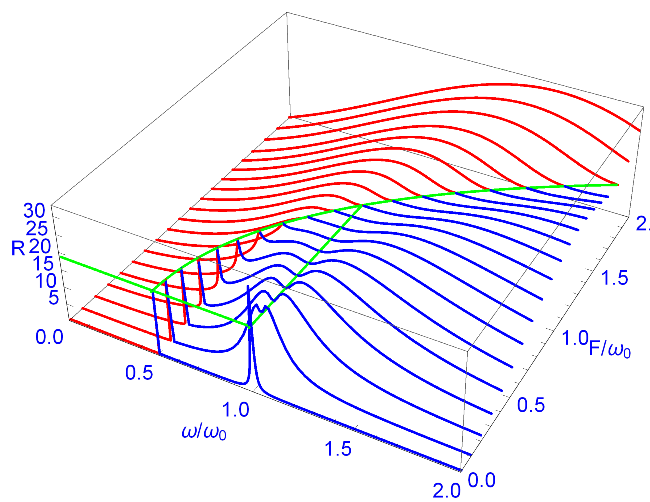

FIG. 7: As Fig. 6, but for $s = 10$. Along the line $\omega = \omega_0$ with $0 < F < \omega_0$ and along the parabola $\omega = \omega_c$ given by Eq. (80) the dissipation rate takes on the constant value $s(s+1)/6 = 55/3$ (green lines). The blue curve with $F/\omega_0 \approx 0$ possesses two unresolved sharp maxima close to an equally sharp minimum at $\omega{\approx}\omega_0$.

as visualized in Figs. 6 and 7 for $s = 1$ and $s = 10$, respectively. For $\frac{1}{2} \leq s \leq \frac{7}{2}$ this value constitutes a smooth maximum for $\omega = \omega_0$ and small F/ω_0 which becomes increasingly sharp for $F/\omega_0 \to 0$. However, for $s > \frac{7}{2}$ the previous maximum turns into a local minimum, the sharpness of which increases for $s \to \infty$. In contrast, along the line $\omega = \omega_c$ this value R_0 remains a local maximum of R for all s, as long as $F < \omega_0$.

For $s \to \infty$ the scaled dissipation rate $r \equiv R/(s^2+s)$ tends to the limit

$$r_\infty(\omega/\omega_0, F/\omega_0) \equiv \frac{\omega}{4\omega_0}\,\frac{F^2}{F^2+(\omega-\omega_0)^2} \tag{117}$$

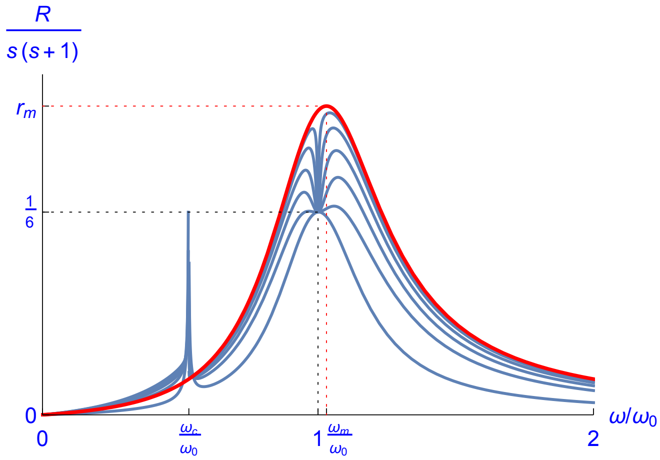

FIG. 8: The scaled dissipation rate $r = R/(s^2+s)$ for $s = \frac{1}{2}$, 5, 10, 20, 50, 200, with bath temperature $\omega_0\beta = 1$ and driving amplitude $F/\omega_0 = 1/4$, as a function of ω/ω_0. The six curves increase with s for $\omega > \omega_c$. They all meet at the two points with coordinates $(\omega_c/\omega_0, 1/6)$ and $(1, 1/6)$. The asymptotic envelope (117) is given by the red curve, with its maximum $(\omega_m/\omega_0, r_m)$ being determined by Eqs. (118) and (119).

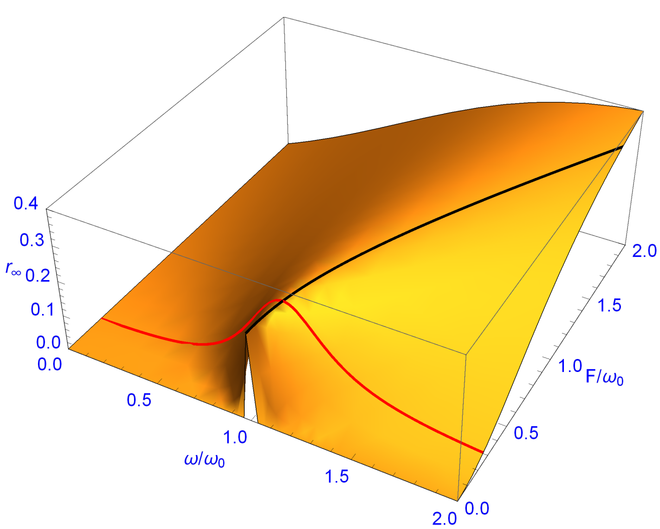

FIG. 9: The asymptotic limit $r_\infty(\omega/\omega_0, F/\omega_0)$ of the scaled dissipation rate $R/(s^2+s)$ for $s \to \infty$ according to Eq. (117). The particular curve with $F/\omega_0 = 1/4$ considered in Fig. 8 is shown in red color. The maximum values of the functions $r_\infty(\omega/\omega_0, F/\omega_0)$ with constant F according to Eqs. (118) and (119) are indicated by a black curve.

which is independent of the heat-bath temperature. As illustrated by Fig. 8 the convergence proceeds pointwise except for $\omega = \omega_c$ and $\omega = \omega_0$ when $F < \omega_0$, where $r = 1/6$ for all s. For fixed F the asymptotic function $r_\infty(\omega/\omega_0, F/\omega_0)$ has a global maximum at

$$\omega_m/\omega_0 = \sqrt{(F/\omega_0)^2 + 1}\,, \qquad (118)$$

adopting the value

$$r_m = \frac{1}{8}\left(1 + \sqrt{(F/\omega_0)^2 + 1}\right)\,, \qquad (119)$$

as depicted in Figs. 8 and 9.

VII. DISCUSSION

A simple harmonic oscillator which is permanently driven by an external time-periodic force while kept in contact with a thermal oscillator bath represents a nonequilibrium system, but nonetheless adopts a steady state and develops a quasistationary distribution of Floquet-state occupation probabilities which equals the Boltzmann distribution of the equilibrium model obtained in the absence of the driving force, being characterized by precisely the same temperature as that of the bath it is coupled to [11, 29].

The system considered in the present work, a spin with arbitrary spin quantum number s exposed to a circularly polarized driving field while interacting with a bath of thermally occupied harmonic oscillators, may be regarded as the next basic model in a hierarchy of analytically solvable models on Periodic Thermodynamics. Exactly as in the case of the linearly forced harmonic oscillator, the system-bath interaction here induces nearest-neighbor coupling among the Floquet states of the time-periodically driven system, so that the model's transition matrix (18) is tridiagonal, thus enforcing detailed balance. Again, the resulting quasistationary Floquet distribution turns out to be Boltzmannian, but now with a quasitemperature which differs from the physical temperature of the bath. Already the mere fact that a time-periodically driven quantum system in its steady state may exhibit a quasitemperature which is different from the actual temperature of its environment, and which can be actively controlled by adjusting, *e.g.*, the amplitude or frequency of the driving force, in itself constitutes a noteworthy observation, suggesting that periodic thermodynamics generally may be far more subtle than usual equilibium thermodynamics based on some effective Floquet Hamiltonian.

Importantly, our model system not only is of basic theoretical interest, but also leads to novel predictions concerning future experiments with paramagnetic materials in strong circularly polarized fields. The very existence of a quasistationary Floquet distribution which is different from the distribution characterizing thermal equilibrium implies that the magnetic response of such a periodically driven material can be quite different from that of the undriven one; as we have demonstrated in Sec. V B, a strong circularly polarized driving field effectively may turn a paramagnetic material into a diamagnetic one. While we are not in a position to ascertain whether the corresponding parameter regime can be reached with already existing experimental set-ups [41], it might be worthwhile to design specifically targeted measurements for confirming this particularly striking prediction of periodic thermodynamics.

Yet, there is still more at stake here. When Brillouin published his now-famous treatise [25] on thermal paramagnetism in 1927, this was essentially a blueprint for an experimental demonstration of the quantization of angular momentum, whereas the further thermodynamical input into the theory was not to be questioned, being backed by the overwhelming generality of equilibrium thermodynamics [27, 28]. At the advent of periodic thermodynamics more than 90 years later, one faces an inverted situation: With the quantization of angular momentum being firmly established, it is nonequilibrium physics in the guise of periodic thermodynamics which is to be examined in measurements of paramagnetism under time-periodic driving. As has been stressed already by Kohn [10] and clarified by Breuer *et al.* [11], quasistationary Floquet distributions are not universal, depending on the very form of the system-bath interaction. Here we have assumed an interaction of the natural-appearing type (11) with coupling (57) proportional to the spin operator S_x on the system's side and simple creation and annihilation operators (13) on the side of the bath, combined with the assumption of a constant spectral density of the bath, but there are other possibilities. Measurements of magnetism under strong driving will be sensitive to such issues; two materials which exhibit precisely the same paramagnetic response in the absence of time-periodic forcing may react differently to a static magnetic field once an additional time-periodic field has been added. Thus, despite the formal similarity of our key results (106) and (107) to their historical antecessors (96) and (97), these former equations may have the potential to open up an altogether new line of research.

Acknowledgments

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