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1997 J. Phys. A: Math. Gen. 30 7193

(http://iopscience.iop.org/0305-4470/30/20/020)

J. Phys. A: Math. Gen. **30** (1997) 7193–7215. Printed in the UK PII: S0305-4470(97)83412-9

On the relation between cavity-dressed states, Floquet states, RWA and semiclassical models

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Received 14 April 1997, in final form 30 June 1997

Abstract. We show that Floquet states can be constructed as strong-field limits of cavity-dressed states. The interaction between laser beams propagating outside the cavity with atoms or molecules are described by Floquet states, constructed from dressed states of average photon number $\bar{n}$ quantized in a cavity of volume V, by taking the limit $V \to \infty$, $\bar{n} \to \infty$, while keeping the photon density $\rho = \bar{n}/V$ constant. Thus, Floquet theory can be seen as a fully quantum-mechanical model in the sense that it describes the photon exchanges between matter and radiation containing a large amount of photons. We discuss in this context adiabatic Floquet theory to treat a slow time dependence of the laser amplitude, to describe pulses, and of its frequency (chirping).

1. Introduction

The control of dynamical processes by intense laser fields is extensively studied in atomic and molecular physics. Efficient tools to treat these phenomena are based on the notion of dressed states, characterizing the stationary states of the molecule dressed by a classical or a quantized radiation field. A method often used to construct dressed states starts with a semiclassical model (i.e. a quantized atom perturbed by a time-dependent classical field), and then invoking the rotating-wave approximation (RWA). This approximation is valid only in the study of a few levels perturbed by a weak and near-resonant field [1]. The new laser sources provide very intense pulsed fields, with the possibility of time-swept frequencies [2–4], for which the RWA is not well adapted, since the resonance conditions for different levels cannot be simultaneously met. In this paper, we present the theory of Floquet states in a formulation that allows us to make a clear connection with the theory of cavity-dressed states [5–7]. We present a construction of Floquet states as large intensity and infinite volume limits of cavity-dressed states. The infinite volume limit is needed in order to take into account the fact that the laser pulse propagates in free space, as opposed to a cavity. Floquet theory gives a precise framework to describe the exchange of a single or a few photons between an intense laser and an atom or molecule. Floquet states can be considered as an intermediate description between quantized cavity-dressed states (which are well adapted to treat intracavity processes) and the semiclassical model. They retain the feature of cavity-dressed states of being able to describe single-photon exchanges, since the considered mode of the radiation field is fully quantized. With the semiclassical model they share the possibility of describing pulses by a (slow) modulation of the coupling amplitude, and to include effects like time-swept frequencies ('chirping'). These phenomena cannot be easily incorporated into cavity-dressed state models, since the intensity of the field is not determined by the coupling constant in the Hamiltonian, but by the initial condition of the quantized radiation field. In the Floquet approach the intensity of the field is completely determined in the Hamiltonian.

Furthermore, we show that the semiclassical model can be recovered from the Floquet approach as a particular form of an interaction representation, together with the choice of coherent states as initial conditions. This construction is related to the analysis of [8–12]. Since the model contains only one (or a few) strongly populated photon modes, it cannot describe spontaneous emission. Its predictions are thus only valid for times that are short compared with the life-times of the relevant molecular or atomic levels.

We also summarize the close connection between the Floquet formalism and the widely used treatment of light-matter interaction with the RWA approach. The essential point of the RWA approach is that the coupling term in the semiclassical Hamiltonian, e.g. for a two-level system coupled with a periodic time-dependent field, is substituted by an approximation which allows us to obtain explicit solutions of the time evolution. These are obtained by first transforming the Schrödinger equation with a time-dependent Hamiltonian into one with a time-independent Hamiltonian. This transformation is unitary, time dependent, and has the same period as the perturbing field. The time-independent Hamiltonian obtained in this way is interpreted in the literature [1] as an effective Hamiltonian containing the information on the atom 'dressed' by the radiation field. This general idea is precisely realized in Floquet theory, and without the need to invoke any approximation (like near resonance or small intensity) [13, 14]. It has been shown in [15, 16] that the Floquet formalism can be alternatively interpreted as a procedure to find a unitary operator that yields an evolution equation with a time-independent Hamiltonian. This unitary transformation can be explicitly expressed in terms of the eigenfunctions of the quasi-energy operator (or Floquet Hamiltonian). Furthermore, the connection with the concepts of cavity-dressed states mentioned above gives a complete picture of dressed states for an atom or molecule interacting in free space with a laser. The shape of the laser pulse and a chirped frequency can be naturally treated by applying adiabatic principles to the Floquet states. The quasi-energies can be represented as a function of the slow time-dependent parameters in quasi-energy diagrams. Quasi-energy diagrams for frequency modulation consist essentially of straight lines, except near avoided crossings where transitions between the dressed states occur [17, 4]. In section 5, we discuss the relation between the slopes of the lines and the relative photon numbers, which provide a useful technique to attribute physical labels to the states.

In appendices A and B we provide mathematical proofs of the main results.

2. Construction of Floquet states from dressed states in a cavity

In a cavity, the dressed states represent the stationary states of an atom or molecule interacting with discrete modes of the quantized electromagnetic radiation [7]. The cavity allows a natural quantization of the radiation, in which, to each mode of frequency ω, there is a corresponding harmonic oscillator of that frequency. Our goal is to study the exchanges of photons between the molecule and the laser field outside the cavity.

2.1. The Floquet theory

The Floquet formalism can be constructed from two different points of view: one approach starts with a semiclassical model and the other one from a completely quantized model in a cavity. We first present the construction of the Floquet formalism from the semiclassical model, and then we establish the relation with the cavity-dressed states. Our formulation allows a direct connection to the phase representation of photon fields.

Our first starting point is a semiclassical model, in which the laser field is described by a classical time-dependent periodic electric field F. The time dependence of the periodic Hamiltonian is introduced through the time evolution of the initial phase $\theta(t) = \theta + \omega t$ [18, 8, 19, 20]:

$$H = H(x, \theta(t)) = H_0 - \mu(x) F(\theta + \omega t) \tag{1}$$

where $\mu(x)$ represents the dipole moment of the molecule. The semiclassical Schrödinger equation

$$\mathrm{i}\hbar \frac{\partial}{\partial t}\varphi = H(x, \theta(t))\varphi \qquad \varphi \in \mathcal{H} \tag{2}$$

is defined on a Hilbert space $\mathcal{H}$, which can be of infinite or finite dimension (e.g. in N-level models $\mathcal{H} = \mathbb{C}^N$). The initial phase θ appears as a parameter. One can think of (2) as a family of equations parametrized by the angle θ. We denote the corresponding family of propagators by $U(t, t_0; \theta)$. The quasi-energy operator K, or Floquet Hamiltonian, is constructed as follows. We define an enlarged Hilbert space

$$\mathcal{K} := \mathcal{H} \otimes \mathcal{L} \tag{3}$$

where $\mathcal{L} := L_2(\mathbb{S}^1, \mathrm{d}\theta/2\pi)$ denotes the space of square integrable functions on the circle $\mathbb{S}^1$ of length 2π. We first lift the family of operators $U(t, t_0; \theta)$ (defined on $\mathcal{H}$) into the operator acting on the enlarged space $\mathcal{K}$, by treating the dependence on θ as a multiplication operator. This operator is unitary in $\mathcal{K}$.

The Floquet Hamiltonian K is then defined as the infinitesimal generator of the following one-parameter $(t - t_0)$ family of unitary operators

$$\mathcal{T}_{-t}\, U(t, t_0; \theta) \mathcal{T}_{t_0} =: \mathrm{e}^{-\mathrm{i}K(\theta)(t-t_0)/\hbar} \equiv U_K(t - t_0, \theta) \tag{4}$$

where the translation operator $\mathcal{T}_t$ acts on $\xi \in L_2(\mathbb{S}^1, \mathrm{d}\theta/2\pi)$, by

$$\mathcal{T}_t \xi(\theta) = \xi(\theta(t)) \tag{5}$$

and can be expressed as

$$\mathcal{T}_t = \mathrm{e}^{\omega t\, \partial/\partial\theta}. \tag{6}$$

We remark that the time evolution of the phase $\theta(t) = \theta + \omega t$ can be seen as a classical flow on the circle $\mathbb{S}^1$ and $\mathcal{T}_t$ is the corresponding Koopman operator [21].

From this definition, the quasi-energy operator, or Floquet Hamiltonian, K, acting on the enlarged space $\mathcal{K}$ can be written as

$$K(\theta) = H(\theta) - \mathrm{i}\hbar\omega \frac{\partial}{\partial\theta}. \tag{7}$$

This formulation leads to the well-established properties of the Floquet states and the quasi-energies (eigenfunctions and eigenvalues of K): stationary states, eigenfunction expansions, etc. We also point out the easy generalization of this formulation to the quasi-periodic case (i.e. the case with several incommensurable frequencies $\underline{\omega} = (\omega_1, \ldots, \omega_d)$, in which case $K = H(\underline{\theta}) - \mathrm{i}\hbar\underline{\omega} \cdot \partial/\partial\underline{\theta}$, with $\underline{\theta} = (\theta_1, \ldots, \theta_d)$ [22–25].

In a second approach we will establish a precise relation between dressed states in a cavity and the Floquet formalism. We show that the Floquet Hamiltonian K can be obtained exactly from the dressed Hamiltonian in a cavity, in the limit of infinite cavity volume and intense laser field. This gives a precise formulation of the statement suggested in [8]: K represents *the dressed Hamiltonian of the molecule interacting in free space with a field* *containing a large number of photons*. Moreover, we establish the physical interpretation of the operator

$$N_r = -i\frac{\partial}{\partial\theta} \tag{8}$$

in the limit of a large number of photons as the *relative photon number operator*. It characterizes the relative photon number of the field with respect to the average $\bar{n}$. The variation of the average of N_r in the Floquet formalism gives the number of photons gained or lost (depending on the sign) by the field.

2.2. Dressed states in intense laser fields

We consider a molecule with Hamiltonian H_0 interacting with one mode of the electromagnetic radiation polarized in the ε direction, where we assume an electric dipole coupling with moment $\boldsymbol{\mu}$ denoting $\mu = \boldsymbol{\mu}\cdot\boldsymbol{\varepsilon}$ [7, 26],

$$H_{\mathrm{LM}} = H_0(x)\otimes \mathbb{1}_{\mathcal{F}} + \mathbb{1}_{\mathcal{H}}\otimes\hbar\omega a^\dagger a - \mu(x)\otimes i\mathcal{E}(a-a^\dagger). \tag{9}$$

The degrees of freedom of the molecule are represented by the variable x and $H_0(x)$ acts on the Hilbert space $\mathcal{H}$. The mode of the laser with frequency ω is described by the number operator of a harmonic oscillator, which can be expressed in terms of the annihilation and creation operators a, $a^\dagger$. They act on the Fock space $\mathcal{F}$ generated by the stationary states $|n\rangle$ of the harmonic oscillator. H_{LM} acts on the enlarged space

$$\mathcal{H}_{\mathrm{LM}} = \mathcal{H}\otimes\mathcal{F}. \tag{10}$$

The coupling constant is given by

$$\mathcal{E} = \sqrt{\frac{\hbar\omega}{2\varepsilon_0 V}} \tag{11}$$

where ε_0 is the vacuum permitivity, and V the volume of the cavity.

We note that, with the cavity-dressed state model (9), the field intensity does not appear explicitly. It depends on the average number of photons contained in the initial state of the field. The connection between this model and the Floquet formulation is given by the following property. Since the radiation is not confined in a cavity, but propagates and interacts with the molecule in free space, we have to take the limit

$V\to\infty$ (infinite cavity volume),

$\bar{n}\to\infty$ (large photon number average),

$\rho = \bar{n}/V =$ constant (constant photon density).

In this limit, the dressed Hamiltonian is identical to the quasi-energy operator K

$$H_{\mathrm{LM}} - \hbar\omega\bar{n} \longrightarrow -i\hbar\omega\frac{\partial}{\partial\theta} + H_0 - \mu E\sin\theta \equiv K \tag{12}$$

where

$$E = \sqrt{\frac{2\rho\hbar\omega}{\varepsilon_0}}. \tag{13}$$

To show this relation, we use the phase representation of H_{LM}, as formulated by Bialynicki–Birula [27–30]. We construct an isomorphism between the Fock space and the space $\mathcal{L}_{\bar{n},\theta}$ defined as a subspace of $\mathcal{L} := L_2(\mathbb{S}^1, \mathrm{d}\theta/2\pi)$, the square integrable periodic functions of the angle θ (i.e. on the circle $\mathbb{S}^1$), generated by the basis functions $\{|e^{ik\theta}\rangle; -\bar{n}\leqslant k < +\infty\}$:

$$|n\rangle\in\mathcal{F}\longleftrightarrow |e^{ik\theta}\rangle\in\mathcal{L}_{\bar{n},\theta} \qquad \text{with } \bar{n}+k=n. \tag{14}$$

In the limit $\bar{n} \to \infty$ we obtain the whole space

$$\mathcal{L}_{\bar{n},\theta} \xrightarrow{\bar{n}\to\infty} L_2\left(\mathbb{S}^1, \frac{\mathrm{d}\theta}{2\pi}\right) \qquad \text{and} \qquad \mathcal{H}_{\mathrm{LM}} \xrightarrow{\bar{n}\to\infty} \mathcal{K} := \mathcal{H} \otimes L_2\left(\mathbb{S}^1, \frac{\mathrm{d}\theta}{2\pi}\right). \tag{15}$$

By this isomorphism, the creation, annihilation and photon-number operators ($a^\dagger$, a and N) have a corresponding representation acting on $\mathcal{L}_{\bar{n},\theta}$, which we denote respectively $a^\dagger_{\bar{n},\theta}$, $a_{\bar{n},\theta}$ and $N_{\bar{n},\theta}$:

$$a^\dagger|n\rangle = \sqrt{n+1}|n+1\rangle \longleftrightarrow a^\dagger_{\bar{n},\theta} = \sqrt{\bar{n} - \mathrm{i}\frac{\partial}{\partial\theta}}\,\mathrm{e}^{\mathrm{i}\theta} P_{\bar{n}} \tag{16a}$$

$$a|n\rangle = \sqrt{n}|n-1\rangle \longleftrightarrow a_{\bar{n},\theta} = \mathrm{e}^{-\mathrm{i}\theta}\sqrt{\bar{n} - \mathrm{i}\frac{\partial}{\partial\theta}}\, P_{\bar{n}} \tag{16b}$$

$$N|n\rangle = a^\dagger a|n\rangle = n|n\rangle \longleftrightarrow N_{\bar{n},\theta} = \left(\bar{n} - \mathrm{i}\frac{\partial}{\partial\theta}\right) P_{\bar{n}} \tag{16c}$$

where $P_{\bar{n}} = \sum_{k=-\bar{n}}^{\infty} |\mathrm{e}^{\mathrm{i}k\theta}\rangle\langle\mathrm{e}^{\mathrm{i}k\theta}|$ is the projector on $\mathcal{L}_{\bar{n},\theta}$. The operators $a^\dagger_{\bar{n},\theta}$, $a_{\bar{n},\theta}$ and $N_{\bar{n},\theta}$ are defined on the whole space $L_2(\mathbb{S}^1, \mathrm{d}\theta/2\pi)$. On the subspace $\mathcal{L}_{\bar{n},\theta}$, the isomorphism can be verified by considering their action on the basis (14). The operator in the coupling term becomes

$$a_{\bar{n},\theta} - a^\dagger_{\bar{n},\theta} = P_{\bar{n}}\left(\mathrm{e}^{-\mathrm{i}\theta}\sqrt{\bar{n} - \mathrm{i}\frac{\partial}{\partial\theta}} - \sqrt{\bar{n} - \mathrm{i}\frac{\partial}{\partial\theta}}\,\mathrm{e}^{\mathrm{i}\theta}\right) P_{\bar{n}} \tag{17}$$

the Hamiltonian is written as

$$H^{(\bar{n})}_{\mathrm{LM}} = H_0(x) \otimes P_{\bar{n}} + 1\!\!1_{\mathcal{H}} \otimes \hbar\omega N_{\bar{n},\theta} - \mu(x) \otimes \mathrm{i}\mathcal{E}(a_{\bar{n},\theta} - a^\dagger_{\bar{n},\theta}). \tag{18}$$

We remark that this is an exact correspondence, which is just a precise expression of Dirac's transformation formalism of quantum mechanics [31, 32].

The explicit writing of the projector $P_{\bar{n}}$ in (16) is motivated by the fact that in this way the operators $H_0(x) \otimes P_{\bar{n}}$, $N_{\bar{n},\theta}$, $a_{\bar{n},\theta}$, $a^\dagger_{\bar{n},\theta}$ and $H^{(\bar{n})}_{\mathrm{LM}}$ are also well defined in the total space $\mathcal{L} = L_2(\mathbb{S}^1, \mathrm{d}\theta/2\pi)$, and the discussion of the limit $\bar{n} \to \infty$ becomes conceptually clearer.

In [27, 28] the formal hypothesis

$$-\mathrm{i}\frac{\partial}{\partial\theta} \ll \bar{n} \tag{19}$$

is invoked to approximate

$$\sqrt{\bar{n} - \mathrm{i}\frac{\partial}{\partial\theta}} = \sqrt{\bar{n}}\sqrt{1 - \frac{\mathrm{i}}{\bar{n}}\frac{\partial}{\partial\theta}} = \sqrt{\bar{n}} + \mathrm{O}\left(\frac{1}{\sqrt{\bar{n}}}\right)$$

which leads to

$$(a_{\bar{n},\theta} - a^\dagger_{\bar{n},\theta})/\sqrt{\bar{n}} \xrightarrow{\bar{n}\to\infty} (\mathrm{e}^{-\mathrm{i}\theta} - \mathrm{e}^{\mathrm{i}\theta}) = -2\mathrm{i}\sin\theta. \tag{20}$$

In the limit $V \to \infty$, $\bar{n} \to \infty$, keeping the photon density $\rho = \bar{n}/V$ constant, we obtain the interaction term

$$\mathrm{i}\mathcal{E}(a_{\bar{n},\theta} - a^\dagger_{\bar{n},\theta}) \longrightarrow \sqrt{\frac{2\rho\hbar\omega}{\varepsilon_0}}\sin\theta. \tag{21}$$

We introduce the laser intensity per unit surface I

$$I = \tfrac{1}{2}\varepsilon_0 c E^2 = \hbar\omega\Phi_{\mathrm{ph}} \tag{22}$$

with the photon velocity c, the field amplitude E and the photon flow $\Phi_{\mathrm{ph}} = \bar{n}c/V$. This allows us to identify the interaction constant of equation (21) with E of equation (12) as $E = \sqrt{2\rho\hbar\omega/\varepsilon_0}$. We obtain thus the Floquet Hamiltonian K of equation (12).

The formal hypothesis (19) must be interpreted in relation with the functions on which $-\mathrm{i}\partial/\partial\theta$ acts. The statement is that if all the states $\{|\mathrm{e}^{\mathrm{i}k\theta}\rangle\}$ that are relevant in the dynamics are such that $|k| \ll \bar{n}$, i.e. if only a few photons are exchanged between light and matter compared to the average photon number $\bar{n}$ contained in the laser field, then *the dressed Hamiltonian* $H_{\mathrm{LM}}^{(\bar{n})}$ *can be identified with the Floquet Hamiltonian* K.

We give in what follows a more precise formulation of this construction based on the dynamics of the coupled system. Since $H_{\mathrm{LM}}^{(\bar{n})}$ and K are both well defined on $\mathcal{H}\otimes L_2(\mathbb{S}^1, \mathrm{d}\theta/2\pi)$, we can compare the time evolutions generated by the two Hamiltonians of any initial state $\psi_0 \in \mathcal{H}\otimes\mathcal{L}$:

Theorem. For N-level models ($\mathcal{H} = \mathbb{C}^N$), given any initial state $\psi_0 \in \mathcal{H}\otimes\mathcal{L}$, there is convergence of the dynamics

$$\lim_{\substack{V,\bar{n}\to\infty\\ \bar{n}/V=\rho}} \mathrm{e}^{-\mathrm{i}(H_{\mathrm{LM}}^{(\bar{n})}/\hbar-\bar{n}\omega)t}\psi_0 = \mathrm{e}^{-\mathrm{i}Kt/\hbar}\psi_0. \tag{23}$$

The detailed statement and the proof of this theorem is given in appendix A.

2.3. Connection with the semiclassical formulation: interaction representation and coherent states

From the formulation of the Floquet formalism given above, we can establish the precise connection between the dynamics in the enlarged space $\mathcal{K}$ defined by the Floquet Hamiltonian K, and the one defined by the semiclassical Hamiltonian in $\mathcal{H}$ with a classical description of the electric field [8]:

The dynamics of the Floquet Hamiltonian in $\mathcal{K}$*, where* θ *is a dynamical variable, is equivalent, in the interaction representation, to the semiclassical Schrödinger evolution in* $\mathcal{H}$*, where* θ *is considered as a parameter corresponding to the fixed initial phase, provided that the initial photon state in the Floquet model is a coherent state.*

For other initial states, as, for example, photon-number states, the Floquet model is not equivalent to the semiclassical model; it contains more structure concerning the photons.

In the enlarged space $\mathcal{K}$, the phase θ of the photons is a quantum-mechanical dynamical variable. It does not have a sharp value in the photon numbers states for example. The uncertainty relations between phase and relative photon number are of the same nature as for the cavity-mode photons, as described in [29] for example. The phase takes a sharp value only for coherent states, as described in sections 2.3.2 and 2.3.4, which allows us to make the connection with the semiclassical model.

2.3.1. Interaction representation. The Schrödinger equation of the Floquet Hamiltonian in $\mathcal{K}$

$$\mathrm{i}\hbar\frac{\partial}{\partial t}\psi(t) = K\psi(t) \tag{24}$$

can be expressed equivalently in an interaction representation defined by the unitary transformation

$$\phi(t) = U_{0\mathrm{r}}^{\dagger}(t)\psi(t) \tag{25}$$

where

$$U_{0r}(t) = e^{-\omega t \partial/\partial\theta} \equiv \mathcal{T}_{-t} \tag{26}$$

is the free photon-field propagator, which we recognize as being identically the Koopman operator (6) used in the Floquet construction of section 2.1. Using equation (4), we obtain

$$\begin{aligned}\phi(t) &= \mathcal{T}_t\psi(t) = \mathcal{T}_t U_K(t-t_0,\theta)\mathcal{T}_{-t_0}\phi(t_0)\\ &= U(t,t_0;\theta)\phi(t_0)\end{aligned}$$

and the evolution equation in this representation becomes

$$i\hbar\frac{\partial}{\partial t}\phi(t) = H(\theta+\omega t)\phi(t) \tag{27}$$

where $\phi(t)\in\mathcal{K}$, i.e. $H(\theta+\omega t)$ is still interpreted as an operator acting on the enlarged Hilbert space $\mathcal{K}$, which, with respect to the variable θ, is a multiplication operator.

Although this equation looks formally like the semiclassical Schrödinger equation (2), we emphasize that it is still different since it is defined in the enlarged Hilbert space $\mathcal{K}$ and the phase θ does not have a definite value, since it is a dynamical variable on the same footing as x. In order to recover the semiclassical equation from (27) we have to reduce it to an equation defined in the Hilbert space $\mathcal{H}$. This can be achieved, as we show in the following, by choosing the initial condition of the photon field as a coherent state.

2.3.2. Coherent states in the limit $\bar{n}\to\infty$. In this section we show that the coherent states are represented in Floquet theory by a generalized function $\Phi_{\theta_0}(\theta)$, which is real, and depends on $\theta-\theta_0$, where $\theta_0\in\mathbb{S}^1$ is a fixed angle, and

$$(\Phi_{\theta_0}(\theta))^2 = 2\pi\delta(\theta-\theta_0). \tag{28}$$

This can be obtained as follows. The photon-field coherent states are eigenvectors of the annihilation operator

$$a|\alpha\rangle = \alpha|\alpha\rangle \qquad \alpha = |\alpha|e^{-i\theta_0}. \tag{29}$$

In the usual Fock-number state representation they are given, up to a phase factor, by

$$|\alpha\rangle = e^{-|\alpha|^2/2}\sum_{n=0}^{\infty}\frac{\alpha^n}{\sqrt{n!}}|n\rangle. \tag{30}$$

In the phase representation they can be written as

$$\Phi_{\theta_0}^{(\bar{n})}(\theta) = e^{i\zeta}e^{-|\alpha|^2/2}\sum_{n=0}^{\infty}\frac{\alpha^n}{\sqrt{n!}}e^{i(n-\bar{n})\theta} = e^{-|\alpha|^2/2}\sum_{n=0}^{\infty}\frac{|\alpha|^n}{\sqrt{n!}}e^{i(n-\bar{n})(\theta-\theta_0)} \tag{31}$$

(where ζ is an arbitrary constant phase that we have chosen as $\zeta=\bar{n}\theta_0$). In order to obtain the representation of coherent states in Floquet theory we have to take $|\alpha|=\sqrt{\bar{n}}$ and then apply the limit $\bar{n}\to\infty$.

This can be rigorously done directly using the representation (31), as we show in appendix B. Here we discuss an alternative construction that is formal but gives a useful intuition. We use an approximate expression of the coherent states for large $\bar{n}$, obtained in [28], by developing

$$a_{\bar{n},\theta} = \sqrt{\bar{n}}e^{-i\theta}\sqrt{1-\frac{1}{\bar{n}}i\frac{\partial}{\partial\theta}} \underset{\bar{n}\to\infty}{\rightsquigarrow} \sqrt{\bar{n}}e^{-i\theta}\left(1-\frac{1}{2\bar{n}}i\frac{\partial}{\partial\theta}\right). \tag{32}$$

This leads to the following asymptotic expression [28] for the normalized coherent state corresponding to $\alpha = \sqrt{\bar{n}}e^{-i\theta_0}$, obtained as a solution of $e^{-i\theta}(1 - i/(2\bar{n})\partial/\partial\theta)\phi_{\theta_0}^{(\bar{n})} = e^{-i\theta_0}\phi_{\theta_0}^{(\bar{n})}$:

$$\Phi_{\theta_0}^{(\bar{n})} \underset{\bar{n}\to\infty}{\rightsquigarrow} \frac{1}{\nu}\exp\{-2\bar{n}[1-\cos(\theta-\theta_0) - i(\sin(\theta-\theta_0) - (\theta-\theta_0))]\} \tag{33}$$

where the normalization constant is

$$\nu^2 = e^{-4\bar{n}} I_0(4\bar{n}) \tag{34}$$

with I_0 a Bessel function, which behaves asymptotically as

$$I_0(4\bar{n}) = \int_0^{2\pi} \frac{d\theta}{2\pi}\exp(4\bar{n}\cos\theta) \underset{\bar{n}\to\infty}{\rightsquigarrow} \frac{e^{4\bar{n}}}{(8\pi\bar{n})^{1/2}}. \tag{35}$$

Therefore

$$|\Phi_{\theta_0}^{(\bar{n})}(\theta)|^2 \underset{\bar{n}\to\infty}{\rightsquigarrow} (8\pi\bar{n})^{1/2}\exp\{-4\bar{n}[1-\cos(\theta-\theta_0)]\} \tag{36}$$

noticing that the function $\exp\{-4\bar{n}[1-\cos(\theta-\theta_0)]\}$ behaves like $\exp\{-2\bar{n}(\theta-\theta_0)^2\}$ for $\bar{n}\to\infty$, we obtain

$$|\Phi_{\theta_0}^{(\bar{n})}(\theta)|^2 \underset{\bar{n}\to\infty}{\rightsquigarrow} 2\pi\delta(\theta-\theta_0) \tag{37}$$

where $\delta(\theta-\theta_0)$ is the analogue on the circle $\mathbb{S}^1$ of the usual Dirac's delta function.

We note that since the phase term in (31) (or in (33)) is odd in $\theta-\theta_0$, we obtain that $\Phi_{\theta_0}^{(\bar{n})}(\theta) \to \Phi_{\theta_0}(\theta)$ with $\Phi_{\theta_0}(\theta)$ real and

$$(\Phi_{\theta_0}^{(\bar{n})}(\theta))^2 \underset{\bar{n}\to\infty}{\longrightarrow} 2\pi\delta(\theta-\theta_0). \tag{38}$$

Furthermore, using the well known properties of the expectation values of N^m on coherent states, we obtain

$$\left\langle \Phi_{\theta_0}^{(\bar{n})}(\theta) \left| -i\frac{\partial}{\partial\theta} \right| \Phi_{\theta_0}^{(\bar{n})}(\theta)\right\rangle_{\mathcal{L}} = 0 \qquad \text{for all } \bar{n} \tag{39}$$

$$\left\langle \Phi_{\theta_0}^{(\bar{n})}(\theta) \left| (-i)^m\frac{\partial^m}{\partial\theta^m} \right| \Phi_{\theta_0}^{(\bar{n})}(\theta)\right\rangle_{\mathcal{L}} \underset{\bar{n}\to\infty}{\longrightarrow} \infty \qquad m \geqslant 2. \tag{40}$$

The subscripts in the scalar product symbols ($\langle\ |\ \rangle_{\mathcal{L}}$) indicate on which space they act. We conclude thus that in Floquet theory the photon coherent states are represented by the 'square root of a δ-function', that we denote by $\Phi_{\theta_0}(\theta) = (2\pi)^{1/2}\delta_{1/2}(\theta-\theta_0)$. Since we will be interested in expectation values, only $|\Phi_{\theta_0}|^2$ will appear in our calculations. In appendix C we discuss some of the formal calculus rules for $\delta_{1/2}(\theta-\theta_0)$.

2.3.3. Expectation values for general initial states of the photon field. For a general initial condition of the photon field $\xi(\theta) \in \mathcal{L}$, we remark that the evolution of the initial condition $\varphi(x)\otimes\xi(\theta)$ can be obtained from the one with the initial condition $\varphi(x)\otimes 1$ (where the constant function $1 \equiv e^{i(k=0)\theta}$ is the relative number state of zero photons):

$$\begin{aligned} U_K(t,\theta)(\varphi(x)\otimes\xi(\theta)) &= \mathcal{T}_{-t}U(t,0;\theta)(\varphi(x)\otimes\xi(\theta)) \\ &= \xi(\theta-\omega t)U(t,0;\theta-\omega t)(\varphi(x)\otimes 1) \\ &= \xi(\theta-\omega t)U_K(t,\theta)(\varphi(x)\otimes 1) \end{aligned} \tag{41}$$

(since $U(t, 0; \theta)$ is a multiplication operator with respect to θ). As a consequence, for any observable $M : \mathcal{K} \to \mathcal{K}$ that with respect to θ is a multiplication operator, we can write the expectation value as

$$\begin{aligned}\langle M\rangle(t) &:= \langle \varphi \otimes \xi | U_K^\dagger(t) M U_K(t) | \varphi \otimes \xi \rangle_{\mathcal{K}} \\ &= \int_0^{2\pi} \frac{\mathrm{d}\theta}{2\pi} |\xi(\theta)|^2 \langle \varphi | U^\dagger(t, 0; \theta) M(\theta + \omega t) U(t, 0; \theta) | \varphi \rangle_{\mathcal{H}} \\ &= \int_0^{2\pi} \frac{\mathrm{d}\theta}{2\pi} |\xi(\theta)|^2 \langle \varphi(t; \theta) | M(\theta + \omega t) | \varphi(t; \theta) \rangle_{\mathcal{H}} \end{aligned} \tag{42}$$

where we denote by $\varphi(t; \theta) \equiv U(t, 0; \theta)\varphi$ the semiclassical evolution with initial phase θ of the initial condition $\varphi \in \mathcal{H}$. In particular, for an observable A of the molecule (i.e. $A \otimes \mathbb{1}_{\mathcal{L}}$) we have

$$\langle A\rangle(t) = \int_0^{2\pi} \frac{\mathrm{d}\theta}{2\pi} |\xi(\theta)|^2 \langle \varphi(t; \theta) | A | \varphi(t; \theta) \rangle_{\mathcal{H}}. \tag{43}$$

2.3.4. Expectation values on coherent states; relation with the semiclassical model. We have stated that we can recover evolution of the semiclassical model from the Floquet evolution in the interaction representation by taking initial states in which the photon field is in a coherent state. This can be formulated more precisely by the following statements. If we take an initial condition of the form $\phi(t = 0) = (2\pi)^{1/2}\varphi(x) \otimes \delta_{1/2}(\theta - \theta_0)$ then

(i) if $A : \mathcal{H} \to \mathcal{H}$ is an observable of the molecule, then according to equation (43)

$$2\pi \langle \varphi \otimes \delta_{1/2}(\theta - \theta_0) | U_K^\dagger(t)(A \otimes \mathbb{1}_{\mathcal{L}}) U_K(t) | \varphi \otimes \delta_{1/2}(\theta - \theta_0) \rangle_{\mathcal{K}} = \langle \varphi(t; \theta_0) | A | \varphi(t; \theta_0) \rangle_{\mathcal{H}}. \tag{44}$$

The last expression is the expectation value calculated with the semiclassical model with initial phase θ_0. We conclude thus that the Floquet evolution with a coherent state in the initial condition is equivalent to the semiclassical model. We note that a somewhat related construction, linking the evolution from cavity-dressed states directly to the semiclassical model (i.e. without the intermediate level of Floquet states as we do here) was established in [10].

(ii) More generally, if $M : \mathcal{K} \to \mathcal{K}$ is an observable that, with respect to θ, is a multiplication operator continuous in θ, then taking for θ a particular value θ_0 defines a family of operators $M(\theta_0) : \mathcal{H} \to \mathcal{H}$, parametrized by θ_0. Then

$$2\pi \langle \varphi \otimes \delta_{1/2}(\theta - \theta_0) | U_K^\dagger(t) M U_K(t) | \varphi \otimes \delta_{1/2}(\theta - \theta_0) \rangle_{\mathcal{K}} = \langle \varphi(t; \theta_0) | M(\theta_0 + \omega t) | \varphi(t; \theta_0) \rangle_{\mathcal{H}}. \tag{45}$$

It was noted in [8, 27] that in the semiclassical model, if the initial phase θ_0 is not known, one can take a statistical average over the initial phases, with uniform distribution:

$$\bar{A}_{\mathrm{sc}} = \int_0^{2\pi} \frac{\mathrm{d}\theta_0}{2\pi} \langle \varphi(t; \theta_0) | A | \varphi(t; \theta_0) \rangle_{\mathcal{H}}. \tag{46}$$

From the discussion above, this coincides with the evolution in the Floquet picture of an initial condition of the photon field that is a photon-number eigenstate $\mathrm{e}^{ik\theta}$ (with arbitrary k). We have seen on the other hand that the semiclassical evolution with an initial phase θ_0 corresponds, in the Floquet picture, to a coherent-state initial condition.

3. Emission and absorption of photons in Floquet theory

In Floquet theory the exchange of photons can be analysed from the temporal variation of the relative photon number. In experiments, one measures for instance the difference in intensity of the laser pulse before and after the interaction with the molecules. We describe this quantity by

$$\delta\langle N\rangle(t) := \left\langle \varphi\otimes\xi \left| U_K^\dagger(t)\left(-\mathrm{i}\frac{\partial}{\partial\theta}\right)U_K(t)\right| \varphi\otimes\xi\right\rangle_{\mathcal{K}} - \left\langle \varphi\otimes\xi \left| -\mathrm{i}\frac{\partial}{\partial\theta}\right| \varphi\otimes\xi\right\rangle_{\mathcal{K}} \tag{47}$$

and we show below that

$$\delta\langle N\rangle(t) = \int_0^{2\pi}\frac{\mathrm{d}\theta}{2\pi\hbar\omega}|\xi(\theta)|^2[\langle\varphi|H(\theta)|\varphi\rangle_{\mathcal{H}} - \langle\varphi(t;\theta)|H(\theta+\omega t)|\varphi(t;\theta)\rangle_{\mathcal{H}}]. \tag{48}$$

In particular, if the photon field is initially in a coherent state $\Phi_{\theta_0}(\theta) = (2\pi)^{1/2}\delta_{1/2}(\theta-\theta_0)$, then

$$\delta\langle N\rangle_{\mathrm{cs}}(t) = \left\langle\varphi\left|\frac{H(\theta_0)}{\hbar\omega}\right|\varphi\right\rangle_{\mathcal{H}} - \left\langle\varphi(t;\theta_0)\left|\frac{H(\theta_0+\omega t)}{\hbar\omega}\right|\varphi(t;\theta_0)\right\rangle_{\mathcal{H}} \tag{49}$$

and, if it is initially in a photon-number eigenstate $|\mathrm{e}^{\mathrm{i}k\theta}\rangle$,

$$\delta\langle N\rangle(t) = \int_0^{2\pi}\frac{\mathrm{d}\theta}{2\pi\hbar\omega}[\langle\varphi|H(\theta)|\varphi\rangle_{\mathcal{H}} - \langle\varphi(t;\theta)|H(\theta+\omega t)|\varphi(t;\theta)\rangle_{\mathcal{H}}]. \tag{50}$$

We remark that $\delta\langle N\rangle(t)$ is independent of the particular k we take, in accordance with the interpretation as the relative photon number.

We can obtain these relations as follows. Using the definition of the quasi-energy operator (7), we can express $\delta\langle N\rangle(t)$ in terms of quantities that do not involve the derivative $-\mathrm{i}\partial/\partial\theta$:

$$\begin{aligned}\delta\langle N\rangle(t) = {} & \left\langle\varphi\otimes\xi\left|U_K^\dagger(t)\frac{K}{\hbar\omega}U_K(t)\right|\varphi\otimes\xi\right\rangle_{\mathcal{K}} \\ & -\left\langle\varphi\otimes\xi\left|U_K^\dagger(t)\frac{H(\theta)}{\hbar\omega}U_K(t)\right|\varphi\otimes\xi\right\rangle_{\mathcal{K}} - \left\langle\varphi\otimes\xi\left|-\mathrm{i}\frac{\partial}{\partial\theta}\right|\varphi\otimes\xi\right\rangle_{\mathcal{K}}.\end{aligned} \tag{51}$$

Using the fact that $[K, U_K] = 0$, $U_K^\dagger U_K = \mathbb{1}$ and equation (7), we can write

$$\delta\langle N\rangle(t) = \left\langle\varphi\otimes\xi\left|\frac{H(\theta)}{\hbar\omega}\right|\varphi\otimes\xi\right\rangle_{\mathcal{K}} - \left\langle\varphi\otimes\xi\left|U_K^\dagger(t)\frac{H(\theta)}{\hbar\omega}U_K(t)\right|\varphi\otimes\xi\right\rangle_{\mathcal{K}} \tag{52}$$

and since

$$\begin{aligned}U_K^\dagger(t,\theta)H(\theta)U_K(t,\theta) &= U^\dagger(t,0;\theta)\mathcal{T}_t H(\theta)\mathcal{T}_{-t}U(t,0;\theta) \\ &= U^\dagger(t,0;\theta)H(\theta+\omega t)U(t,0;\theta)\end{aligned} \tag{53}$$

we obtain equation (48).

We can also obtain more precise information on the probability $P(L,t)$ that L photons are exchanged. If at time $t=0$ the photon field is in a photon-number eigenstate $\mathrm{e}^{\mathrm{i}k\theta}$ and $\psi(t=0)=\psi_0=\varphi\otimes\mathrm{e}^{\mathrm{i}k\theta}$ then the probability that a measurement performed at time t yields that L photons have been exchanged, is given by

$$\begin{aligned}P(L,t) &= \langle U_K(t)\psi_0|[\mathbb{1}_{\mathcal{H}}\otimes|\mathrm{e}^{\mathrm{i}(L+k)\theta}\rangle\langle\mathrm{e}^{\mathrm{i}(L+k)\theta}|]|\; U_K(t)\psi_0\rangle_{\mathcal{K}} \\ &= \sum_n |\langle\varphi_n\otimes\mathrm{e}^{\mathrm{i}(k+L)\theta}|U_K(t)(\varphi\otimes\mathrm{e}^{\mathrm{i}k\theta})\rangle_{\mathcal{K}}|^2\end{aligned} \tag{54}$$

where $\{\varphi_n\}$ is an arbitrary basis of $\mathcal{H}$.

3.1. Invariance with respect to the choice of the origin of the relative photon number

Due to the relative character of the number operator $-\mathrm{i}\partial/\partial\theta$, all the physical predictions of the Floquet model must be invariant with respect to a global translation of the relative photon numbers. We show that this is indeed the case for the properties discussed in the previous section.

The probability $P(L,t)$ is independent of the particular initial photon-number state chosen, i.e. it is independent of k since:

$$U_K(t)(\varphi\otimes \mathrm{e}^{\mathrm{i}k\theta}) = U(t,0;\theta-\omega t)(\varphi\otimes \mathrm{e}^{\mathrm{i}k(\theta-\omega t)}) \tag{55}$$

and thus

$$P(L,t) = \sum_n |\langle\varphi_n\otimes \mathrm{e}^{\mathrm{i}L\theta}|U_K(t)(\varphi\otimes 1)\rangle_{\mathcal{K}}|^2. \tag{56}$$

For the average number of exchanged photons $\delta\langle N\rangle(t)$ it is straightforward to verify that one obtains the same result for the choice of any initial condition of the photon field of the form

$$\xi = \sum_k c_k \mathrm{e}^{\mathrm{i}(k+m)\theta} \tag{57}$$

with arbitrary translation m.

3.2. Algorithmic aspects

From the relation (4), it follows that the information contained in the Floquet evolution can be obtained from the numerical simulation of the semiclassical model, and vice versa. Indeed, performing one simulation of the Floquet evolution in $\mathcal{K}$ contains the same information as a family of simulations of the semiclassical model in $\mathcal{H}$ for different values of the initial phase θ. (A finite number N of semiclassical simulations corresponds to one Floquet simulation with N grid points in the discretization of θ.) A single semiclassical simulation yields the Floquet evolution corresponding to a photon coherent state in the initial condition. Extended Hilbert-space techniques of this type have been applied to the numerical solution of the Schrödinger equation in [33].

4. Structure of the Floquet states; relation with RWA dressed states

The widely used RWA (see e.g. [34, 1]) allows us to obtain an approximation of the solution of the time-dependent Schrödinger equation analytically for a two-level system driven periodically (extensions to N-level systems have been developed, under some particular conditions on the spacings [35] of the levels). In this section we will set $\hbar = 1$. For a Hamiltonian of the form

$$H(\theta+\omega t) = \frac{\omega_0}{2}\sigma_z + \Omega\sin(\theta+\omega t)\sigma_y \tag{58}$$

with the Pauli matrices $\sigma_x, \sigma_y, \sigma_z$, the RWA consists in substituting the interaction $\Omega\sin(\theta+\omega t)\sigma_y$ by

$$\frac{\Omega}{2}\begin{pmatrix} 0 & \mathrm{e}^{-\mathrm{i}(\theta+\omega t)} \\ \mathrm{e}^{\mathrm{i}(\theta+\omega t)} & 0 \end{pmatrix}.$$

This comes down to keeping the term oscillating with ω and neglecting the term oscillating with $-\omega$, which produces a counter-rotating correction to the RWA. The RWA is only justified when the driving frequency is resonant or weakly detuned (i.e. $\omega \simeq \omega_0$).

Furthermore, the RWA gives a good approximation only if the coupling (i.e. the Rabi frequency Ω) is small compared with the Bohr frequency ω_0. In this sense, the RWA is a near-resonance, weak-field approximation [1]. Otherwise, nonlinear effects, like the dynamical Stark effect, destroy the near-resonance property. Once this approximation is made, the time evolution can be solved explicitly. The idea is to find a unitary time-dependent transformation (with the same period as the perturbation field) which leads to an equation with a time-independent Hamiltonian. The point that we want to stress in this section is that, within the approximations considered above, dressed states of the RWA Hamiltonian are just approximations of the Floquet states [36, 1]. Floquet states are indeed well defined for the complete Hamiltonian (58), and they play the same role as the RWA dressed states, but without the need of approximation. As it was shown by Salzman [15] finding a unitary operator that transforms the Schrödinger equation with a time-dependent Hamiltonian into one with a time-independent Hamiltonian is equivalent with the spectral problem of the Floquet Hamiltonian. In this sense, the Floquet theory can be thought of as a generalization of the RWA, but without using any approximation.

This connection can be seen from the following alternative interpretation of the quasi-energy eigenvalue problem [15], which gives information about the general structure of the spectral elements of the quasi-energy operator K. We look for a unitary transformation $C(x,\theta) : \mathcal{H} \to \mathcal{H}$ (θ is treated here as a parameter) to get a time-independent Schrödinger equation, i.e. such that

$$U^B(t, t_0; \theta) = C(\theta(t))^{-1} U(t, t_0; \theta) C(\theta(t_0)) \tag{59}$$

satisfies

$$\mathrm{i}\frac{\partial}{\partial t} U^B(t, 0; \theta) = B U^B(t, 0; \theta) \tag{60}$$

where $B(x)$ is a time- and θ-independent operator acting on $\mathcal{H}$. We remark that there always exists a unitary time-dependent transformation that leads to an equation with a time-independent Hamiltonian. But here we require specifically that the unitary transformation C depends on time only through the variable $\theta(t)$, i.e. that it is periodic with the same frequency as the perturbation.

With such a transformation we have

$$U(t, t_0; \theta) = C(\theta(t)) \mathrm{e}^{-\mathrm{i}B(t-t_0)} C(\theta(t_0))^{-1} \tag{61}$$

with

$$B = C(\theta(t))^{-1} H(\theta(t)) C(\theta(t)) - \mathrm{i} C(\theta(t))^{-1} \frac{\partial C(\theta(t))}{\partial \theta(t)} \frac{\mathrm{d}\theta(t)}{\mathrm{d}t}. \tag{62}$$

Acting with $\mathcal{T}_{-t}$ from the left on equation (61), and with $\mathcal{T}_{t_0}$ from the right, we show that the unitary transform C induces a unitary transform of the quasi-energy operator in the enlarged space $\mathcal{K}$

$$\mathrm{e}^{-\mathrm{i}K(\theta)(t-t_0)} = C(\theta) \mathcal{T}_{-t} \mathrm{e}^{-\mathrm{i}B(t-t_0)} \mathcal{T}_{t_0} C(\theta)^{-1}. \tag{63}$$

Thus, finding the spectral elements of $\mathrm{e}^{-\mathrm{i}K(\theta)t}$ in $\mathcal{K}$ comes down to determining eigenvalues and eigenfunctions of the time-independent operator B in $\mathcal{H}$ (denoted respectively by λ_m^B and Ψ_m^B, $m \in \mathbb{N}$) and those of the Koopman operator $\mathcal{T}_{-t}$ in $L_2(\mathbb{S}^1, \mathrm{d}\theta/2\pi)$, $\mathrm{e}^{-\mathrm{i}k\omega t}$ and $\mathrm{e}^{\mathrm{i}k\theta}$, $k \in \mathbb{Z}$, and then apply the transform (63).

From this we can deduce the general structure of the Floquet states

$$\Psi_{m,k}(x, \theta) = C(x, \theta)[\mathrm{e}^{\mathrm{i}k\theta} \otimes \Psi_m^B(x)] \tag{64}$$

and of the quasi-energy spectrum

$$\lambda_{m,k} = \lambda_m^B + k\omega. \tag{65}$$

For the RWA Hamiltonian, the transformation is

$$C(\theta) = \begin{pmatrix} 1 & 0 \\ 0 & \mathrm{e}^{\mathrm{i}\theta} \end{pmatrix} \tag{66}$$

which leads to the constant operator (62) $B = (\omega_0 - \omega)\sigma_z/2 + \Omega\sigma_x/2 + \omega\mathbb{1}/2$, which yields the RWA dressed states.

5. Adiabatic Floquet theory for the chirped laser field and labelling of the dressed states

The shape of the pulse and a swept frequency can be modelled by adding into the periodic Hamiltonian a slow time dependence compared with the period. This can be treated by adiabatic techniques and Landau–Zener-type formulae to study transitions between levels. Breuer *et al* [38] have treated the case of a slow time-dependent amplitude. Its study involves quasi-energy diagrams representing the quasi-energies as a function of the amplitude. This has been applied successfully to control population transfer to enhance tunnelling time [39, 40], as well as to interpret a large variety of experiments [41–46]. The adiabatic Floquet theory for a more general slow time dependence, including a frequency modulation in addition to the amplitude modulation, has been developed in [4]. The theory establishes the determining role of an effective frequency driving the dynamics of the system, and which is the relevant parameter as a function of which one has to study quasi-energy diagrams. Here we will discuss the use of the relative photon number operator N_r to assign physical labels to the states in quasi-energy diagrams.

We consider a Hamiltonian $H^{[r(t)]}(\theta + \omega(t)t)$ where $r(t)$ represents a set of time-dependent parameters, not including the swept frequency $\omega(t)$. The solution of the time-dependent Schrödinger equation is related to the propagator associated to an instantaneous quasi-energy operator which is defined through an effective frequency.

The operator U is the propagator of the Schrödinger equation

$$\mathrm{i}\hbar\frac{\partial}{\partial t}U(t, t_0; \theta) = H^{[r(t)]}(\theta + \omega(t)t)U(t, t_0; \theta) \tag{67}$$

if and only if the operator U_K, defined by

$$U_K(t, t_0, \theta) = \mathcal{T}_{-t}U(t, t_0; \theta)\mathcal{T}_{t_0} \tag{68}$$

satisfies

$$\mathrm{i}\hbar\frac{\partial}{\partial t}U_K(t, t_0, \theta) = K^{[r(t), \omega_{\mathrm{eff}}(t)]}(\theta)U_K(t, t_0, \theta) \tag{69}$$

where $\omega_{\mathrm{eff}}(t) = \omega(t) + \dot{\omega}(t)t$, $\mathcal{T}_t$ is the translation operator which acts on $L_2(\mathbb{S}^1, \mathrm{d}\theta/2\pi)$ as $\mathcal{T}_t\xi(\theta) = \xi(\theta + \omega(t)t)$ and

$$K^{[r(t), \omega_{\mathrm{eff}}(t)]}(\theta) = H^{[r(t)]}(\theta) - \mathrm{i}\hbar\omega_{\mathrm{eff}}(t)\frac{\partial}{\partial\theta}. \tag{70}$$

This result is proved as follows. We start with equation (69) which, by differentiation of (68) and the fact that for any $\xi \in \mathcal{L}$

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}(\mathcal{T}_{-t}\xi(\theta)) &= \frac{\mathrm{d}}{\mathrm{d}t}\xi(\theta - \omega(t)t) = -\frac{\mathrm{d}\theta}{\mathrm{d}t}\mathcal{T}_{-t}\frac{\partial}{\partial\theta}\xi(\theta) \\ &= -(\omega + t\dot{\omega})\mathcal{T}_{-t}\frac{\partial}{\partial\theta}\xi(\theta) = -\omega_{\mathrm{eff}}\mathcal{T}_{-t}\frac{\partial}{\partial\theta}\xi(\theta)\end{aligned} \tag{71}$$

is equivalent to

$$-\mathrm{i}\hbar\omega_{\text{eff}}\mathcal{T}_{-t}\frac{\partial}{\partial\theta}U\mathcal{T}_{t_0}+\mathrm{i}\hbar\mathcal{T}_{-t}\frac{\partial}{\partial t}U\mathcal{T}_{t_0}=K^{[r,\omega_{\text{eff}}]}\mathcal{T}_{-t}U\mathcal{T}_{-t_0}. \tag{72}$$

or also to,

$$\begin{aligned}-\mathrm{i}\hbar\omega_{\text{eff}}\frac{\partial}{\partial\theta}U+\mathrm{i}\hbar\mathcal{T}_{-t}\frac{\partial}{\partial t}U &= \mathcal{T}_t K^{[r,\omega_{\text{eff}}]}\mathcal{T}_{-t}U\\ &= -\mathrm{i}\hbar\omega_{\text{eff}}\frac{\partial}{\partial\theta}U+\mathcal{T}_{-t}H^{[r]}\mathcal{T}_{-t}U\end{aligned} \tag{73}$$

which is identical to the Schrödinger equation:

$$\mathrm{i}\hbar\frac{\partial}{\partial t}U(t,t_0;\theta)=H^{[r(t)]}(\theta(t))U(t,t_0;\theta). \tag{74}$$

The appearance of an *effective* instantaneous frequency in the quasi-energy operator distinguishes our formulation from earlier attempts to treat adiabatic frequency modulation [38]. Extending the usual adiabatic theorem to the instantaneous Floquet states, one can formulate under suitable conditions the following adiabatic principle.

If at time t_0 the system is an instantaneous Floquet state, then in the adiabatic limit the time evolution $\phi(t)$ stays for all t in the connected instantaneous Floquet eigenstate:

$$\phi(t)=\mathrm{e}^{-\mathrm{i}\delta_m(t)}\Psi_m^{[r(t),\omega_{\text{eff}}(t)]}(\theta+\omega(t)t) \tag{75}$$

where the phase $\delta_m\in\mathbb{R}$ is the superposition of the dynamical phase and Berry's geometric phase [47].

This formulation leads to the analysis of the quasi-energy operator $K^{[r,\omega_{\text{eff}}]}$ (70), which is evaluated at each (fixed) value of the parameters r and ω_{eff}.

The limitation of the adiabatic behaviour occurs around avoided crossings between the quasi-energy levels, which produce transitions between these levels. This can be calculated with Landau–Zener formula in an adiabatic regime, in the sense that the subspace generated by the quasi-energies involved in the avoided crossings has an adiabatic evolution with respect to the other states.

We can calculate the average of $N_{\mathrm{r}}=-\mathrm{i}\partial/\partial\theta$ in a Floquet state Ψ_n by differentiating:

$$\begin{aligned}\frac{\partial\lambda_n}{\partial\omega_{\text{eff}}}&=\frac{\partial}{\partial\omega_{\text{eff}}}\langle\Psi_n|K|\Psi_n\rangle_{\mathcal{K}}\\&=\left\langle\Psi_n\left|-\mathrm{i}\hbar\frac{\partial}{\partial\theta}\right|\Psi_n\right\rangle_{\mathcal{K}}+\left\langle\frac{\partial\Psi_n}{\partial\omega_{\text{eff}}}\left|K\right|\Psi_n\right\rangle_{\mathcal{K}}+\left\langle\Psi_n\left|K\right|\frac{\partial\Psi_n}{\partial\omega_{\text{eff}}}\right\rangle_{\mathcal{K}}\end{aligned}$$

using the fact that $H(\theta)$ does not depend on ω_{eff}. From the relation $\langle\Psi_n|\Psi_n\rangle=1$, we deduce

$$\left\langle\frac{\partial\Psi_n}{\partial\omega_{\text{eff}}}\left|K\right|\Psi_n\right\rangle_{\mathcal{K}}+\left\langle\Psi_n\left|K\right|\frac{\partial\Psi_n}{\partial\omega_{\text{eff}}}\right\rangle_{\mathcal{K}}=\lambda_n\left(\frac{\partial}{\partial\omega_{\text{eff}}}\langle\Psi_n|\Psi_n\rangle\right)=0 \tag{76}$$

which finally gives the identity

$$\langle\Psi_n|N_{\mathrm{r}}|\Psi_n\rangle_{\mathcal{K}}=\frac{1}{\hbar}\frac{\partial\lambda_n}{\partial\omega_{\text{eff}}}. \tag{77}$$

This gives the general behaviour of the quasi-energy diagram as a function of the effective frequency: *the quasi-energy diagram as a function of the effective frequency is essentially composed of straight lines, except where the quasi-energies form avoided crossings at which there are transitions between the levels*.

This can be seen, for example, on a quasi-energy diagram for a three-level ladder system [2]. We justify this property using the physical interpretation of the relative photon number operator applied to the relation (77): far from any avoided crossing, no transition occurs, i.e. there is no variation of the average relative photon number in a given Floquet state: $\partial\lambda_n/\partial\omega_{\text{eff}}$ is constant, i.e. $\lambda_n(\omega_{\text{eff}})$ is a straight line. This implies the exchange of slope of the two quasi-energies around an avoided crossing. Instead of labelling the quasi-energies by continuity as it is usually done (e.g. to apply the Landau–Zener formula), we can label them with respect to their slope, which reflects the effective transition that takes place between the quasi-energies at the avoided crossings.

Acknowledgments

Support from the EC contract ERBCHRXCT94-0460 for the project 'Stability and universality in classical mechanics', from grants of the Conseil Régional de Bourgogne and the Commissariat à l'Energie Atomique are acknowledged.

Appendix A. Proof of the convergence from cavity to Floquet dynamics

In this appendix, we will show that in the limit when the mean number of photons tends to infinity, while keeping their density constant, the dressed energy operator is equal to the Floquet Hamiltonian. To keep the argument simple, we will treat only the case $\mathcal{H} = \mathbb{C}^N$. As it can readily be seen from the proofs, all the statements below remain valid if μ and H_0 are merely bounded symmetric operators on a Hilbert space $\mathcal{H}$.

To simplify the notation, we will let $\hbar = 1$ and use the following:

$$\begin{aligned}
V_{\bar{n}} &= -\mathrm{i}\frac{E}{\sqrt{\bar{n}}}\left(\mathrm{e}^{-\mathrm{i}\theta}\sqrt{\bar{n} - \mathrm{i}\frac{\partial}{\partial\theta}} - \sqrt{\bar{n} - \mathrm{i}\frac{\partial}{\partial\theta}}\,\mathrm{e}^{\mathrm{i}\theta}\right)P_{\bar{n}} \\
V &= -2E\sin\theta \\
K_0 &= -\mathbb{1}\otimes \mathrm{i}\omega\frac{\partial}{\partial\theta} + H_0\otimes\mathbb{1} \\
H_{\bar{n}} &:= H_{\text{LM}}^{(\bar{n})} - \omega\bar{n} = -\mathbb{1}\otimes\mathrm{i}\omega\frac{\partial}{\partial\theta}P_{\bar{n}} + H_0\otimes P_{\bar{n}} + \mu\otimes V_{\bar{n}} =: K_0P_{\bar{n}} + W_{\bar{n}} \\
K &= -\mathbb{1}\otimes\mathrm{i}\omega\frac{\partial}{\partial\theta} + H_0\otimes\mathbb{1} + \mu\otimes V =: K_0 + W
\end{aligned} \tag{A1}$$

where we have denoted by $P_{\bar{n}} = \sum_{k\geqslant -\bar{n}} |\mathrm{e}^{\mathrm{i}k\theta}\rangle\langle\mathrm{e}^{\mathrm{i}k\theta}|$ the eigenprojection of $-\mathrm{i}\partial/\partial\theta$ on the eigenspace of Fourier modes $\geqslant -\bar{n}$. Our aim is to prove the following result, which shows that the evolutions associated to K and $H_{\bar{n}}$ are equal in the limit $\bar{n}\to\infty$.

Theorem A1. Let H_0 and μ be symmetric matrices (or bounded self adjoint operators) on $\mathcal{H}$. Then, $\mathrm{e}^{-\mathrm{i}tH_{\bar{n}}}$ converges strongly to $\mathrm{e}^{-\mathrm{i}tK}$ for all $t\in\mathbb{R}$, as $\bar{n}\longrightarrow\infty$, i.e.

$$\mathrm{e}^{-\mathrm{i}tH_{\bar{n}}}\psi \longrightarrow \mathrm{e}^{-\mathrm{i}tK}\psi \qquad \text{for all } \psi\in\mathcal{K} = \mathcal{H}\otimes L_2(\mathbb{S}^1, \mathrm{d}\theta/2\pi). \tag{A2}$$

Note that this is the best one can hope for the kind of problems we are concerned with, as the convergence cannot be uniform.

The idea of the proof is rather straightfoward and simple. We will show that it is sufficient to verify that $H_{\bar{n}}\psi$ converges to $K\psi$ for all vectors ψ of the form $\varphi\otimes\mathrm{e}^{\mathrm{i}k\theta}$ with

$\varphi \in \mathcal{H}$, to insure that the corresponding evolutions are identical in the limit. This is a simple consequence of the fact that, provided $\bar{n} \geqslant -k$:

$$H_{\bar{n}}\varphi \otimes \mathrm{e}^{\mathrm{i}k\theta} = K_0\varphi \otimes \mathrm{e}^{\mathrm{i}k\theta} - \mathrm{i}E(\mu\varphi) \otimes \left(\sqrt{1+\frac{k}{\bar{n}}}\mathrm{e}^{-\mathrm{i}\theta} - \sqrt{1+\frac{k+1}{\bar{n}}}\mathrm{e}^{\mathrm{i}\theta}\right)\mathrm{e}^{\mathrm{i}k\theta}. \tag{A3}$$

However, to prove theorem A1, we have to deal with unbounded operators for which one always needs to be cautious, as it is necessary to deal with problems related to the domain of the operator and notions of generalized convergence. The questions associated to convergence can be discussed by considering a set of bounded functions of the unbounded self adjoint operators involved in the limit procedure. Natural families of such bounded functions are given by 'propagators' $\{u_t(x) = \mathrm{e}^{\mathrm{i}tx}; t \in \mathbb{R}\}$ and 'resolvents' $\{r_z(x) = (x-z)^{-1}; \Im mz \neq 0\}$.

In the proof, we will need several classical results in operator theory that can be found in the series of Reed and Simon [48] or in the book of Kato [49]. For the convenience of the reader we will, however, state those results when needed.

First, recall that $\mathrm{dom}\, T$, the *domain* of an (unbounded) operator T, is the set where it has been defined. By definition, $\psi \in \mathrm{dom}\, T$ implies that $\|T\psi\| < \infty$. An operator S is said to be an *extension* of an operator T, if $\mathrm{dom}\, T \subset \mathrm{dom}\, S$ and $S\psi = T\psi$ for each $\psi \in \mathrm{dom}\, T$. This will be denoted by $T \subset S$.

The notion of general convergence we will use is the following. A sequence of self adjoint operators $\{T_n\}$ is said to *converge strongly in the resolvent sense* to a self adjoint operator T, if for any z with $\Im mz \neq 0$, the sequence $(T_n - z)^{-1}\psi$ converges to $(T-z)^{-1}\psi$ for all vectors ψ. The most natural convergence for our problem would have been to directly consider the propagators. But, the chosen one allows us to rely on the results found in [48]. In fact, these two kinds of convergence are equivalent, as shown by the following result due to Trotter [48, theorem VIII.21, p 287].

Theorem A2 (Trotter). Let $\{T_n\}$ and T be self adjoint operators. Then $T_n \longrightarrow T$ in the strong resolvent sense if and only if $\mathrm{e}^{\mathrm{i}tT_n}$ converges strongly to $\mathrm{e}^{\mathrm{i}tT}$ for each t.

In order to prove the convergence of $H_{\bar{n}}$ to K in the strong resolvent sense, we need to introduce the notion of essential self adjointedness. First, recall that an operator T is said to be *closed*, if it satisfies the following property: if $\{\psi_n\} \subset \mathrm{dom}\, T$ converge to ψ and if $T\psi_n \longrightarrow \varphi$, then this implies that $\psi \in \mathrm{dom}\, T$ and $\varphi = T\psi$. An operator T is called *closable*, if it admits a closed extension. The *closure* of a closable operator is the smallest closed extention of this operator (which exists by hypothesis).

If T has a dense domain, we can define its *adjoint* $T^\dagger$ by

$$\langle T^\dagger\psi|\varphi\rangle = \langle\psi|T\varphi\rangle \qquad \text{for all } \varphi \in \mathrm{dom}\, T \tag{A4}$$

and its domain $\mathrm{dom}\, T^\dagger$ is the set of vectors ψ such that there exists a constant $C = C_\psi$ with $|\langle\psi|T\varphi\rangle| \leqslant C\|\varphi\|$, for all $\varphi \in \mathrm{dom}\, T$. It can be proven that $T^\dagger$ is always closed (see [48, theorem VIII.1, p 252]).

A *symmetric* operator T is an operator satisfying $T \subset T^\dagger$, and hence it is closable. An operator is called *self adjoint*, if it is equal to its adjoint. This implies in particular that their domains are equal. A symmetric operator is *essentially self adjoint*, if its closure is self adjoint. In general, it is very difficult to prove that a symmetric operator is self adjoint, as the domain of a self adjoint operator is not easy to find. So one deals with symmetric operators and tries to prove that they are essentially self adjoint.

The basic criterion for essential self adjointness is the corollary following theorem VIII.3 in [48, p 257].

Proposition A3. Let T be a symmetric operator. Then the following are equivalent:

(a) T is essentially self adjoint;
(b) $\mathrm{kernel}(T^\dagger \pm \mathrm{i}) = \{0\}$;
(c) $\mathrm{range}(T \pm \mathrm{i})$ are dense.

As a useful illustration of this result, we will prove the following.

Lemma A4. A symmetric operator T having a total set of eigenvectors is essentially self adjoint on the set of all finite linear combinations of its eigenvectors.

Indeed, by symmetry all the eigenvalues λ_j of T are real, hence the set $\{(T \pm \mathrm{i})\varphi_n = (\lambda_n \pm \mathrm{i})\varphi_n \neq 0$ is total, which implies that the finite linear combinations are dense in the Hilbert space.

The next result is a reformulation of [48, theorem VIII.25, p 292], which shows that it is sufficient to prove that a sequence of self adjoint operators converges strongly to a self adjoint operator T on a set D on which there are all essentially self adjoint†.

Theorem A5. Let $\{T_n\}$ and T be essentially self adjoint on a common domain D. If $T_n\varphi \longrightarrow T\varphi$ for each $\varphi \in D$, then $T_n \longrightarrow T$ in the strong resolvent sense.

In order to use this result, we need to show that $H_{\bar{n}}$ and K are essentially self adjoint on a common domain. This domain will be the set of finite linear combinations of simple tensors of the form $\varphi \otimes \mathrm{e}^{\mathrm{i}k\theta}$:

$$\mathcal{D} = \left\{ \sum_{\text{finite}} a\varphi \otimes \mathrm{e}^{\mathrm{i}k\theta}; \varphi \in \mathcal{H} \right\}. \tag{A5}$$

We first have to deal with the tensorial form of the operators involved. We can use the corollary following theorem VIII.33 in [48, p 301].

Proposition A6. Let $T_1, \ldots, T_N$ be self adjoint operators on $\mathcal{H}_1, \ldots, \mathcal{H}_N$ and suppose that for each k, D_k is a domain of essential self adjointness for T_k. Then, the operators $T_1 \otimes \ldots \otimes T_N$ and $T_1 \otimes \mathbb{1} \otimes \ldots \otimes \mathbb{1} + \ldots + \mathbb{1} \otimes \ldots \otimes \mathbb{1} \otimes T_n$ are essentially self adjoint on $D = \otimes_{k=1}^N D_k$, the set of finite linear combinations of simple tensors.

This proposition shows that $H_0 \otimes P_{\bar{n}}$, $H_0 \otimes \mathbb{1}$ and $-\mathbb{1} \otimes \mathrm{i}\omega\partial/\partial\theta$ are essentially self adjoint on $\mathcal{D}$ and hence they are in particular symmetric.

Next, to prove that $H_{\bar{n}}$ and K are indeed essentially self adjoint on $\mathcal{D}$, we will need the following result that can be found in [49, chapter V.4, theorem 4.6, p 289].

Theorem A7. Let T be essentially self adjoint and let A be symmetric. If A satisfies

$$\|A\psi\|^2 \leqslant a^2\|\psi\|^2 + b^2\|T\psi\|^2 \qquad \text{for all } \psi \in \operatorname{dom} T \tag{A6}$$

with $b \leqslant 1$, then $T + A$ is essentially self adjoint on $\operatorname{dom} T$.

Operators satisfying (A6) are said T-bounded, with relative T-bound $b \leqslant 1$.

$H_0 \otimes P_{\bar{n}}$, $H_0 \otimes \mathbb{1}$ and $W = -2E\mu \otimes \sin\theta$ being a bounded operator, they satisfy (A6) with $b = 0$ with respect to any essentially self adjoint operator. This proves that K is essentially self adjoint on $\mathcal{D}$. It remains to see that

$$W_{\bar{n}} = \mu \otimes V_{\bar{n}} = -\mathrm{i}\mu \otimes \frac{E}{\sqrt{\bar{n}}} \left(\mathrm{e}^{-\mathrm{i}\theta}\sqrt{\bar{n} - \mathrm{i}\frac{\partial}{\partial\theta}} - \sqrt{\bar{n} - \mathrm{i}\frac{\partial}{\partial\theta}}\mathrm{e}^{\mathrm{i}\theta} \right) P_{\bar{n}} \tag{A7}$$

† For the reformulation, see the definition of a *core* in [48, chapter VIII.2, p 256].

is $-\mathbb{1}\otimes \mathrm{i}\omega\partial/\partial\theta P_{\bar n}$-bounded with relatively bound $\leqslant 1$ on the set $\mathcal{D}$, as we can then add the bounded operator $H_0\otimes P_{\bar n}$ without changing this quality. First, note that

$$V_{\bar n}\mathrm{e}^{\mathrm{i}k\theta} = -\mathrm{i}\frac{E}{\sqrt{\bar n}}\left(\sqrt{\bar n+k}\mathrm{e}^{-\mathrm{i}\theta} - \sqrt{\bar n+k+1}\mathrm{e}^{\mathrm{i}\theta}\right)P_{\bar n}\mathrm{e}^{\mathrm{i}k\theta} \qquad \text{for any } k. \tag{A8}$$

Using the fact that $W_{\bar n} = \mu\otimes V_{\bar n}$ can be written as $(\mu\otimes\mathbb{1})(\mathbb{1}\otimes V_{\bar n})$ and that the multiplication by $\mathrm{e}^{\pm\mathrm{i}\theta}$ is an unitary operator on $\mathcal{K}$, we have for any vector $\sum\varphi_k\otimes\mathrm{e}^{\mathrm{i}k\theta}$ in $\mathcal{D}$ that†,

$$\begin{aligned}\left\|W_{\bar n}\sum\varphi_k\otimes\mathrm{e}^{\mathrm{i}k\theta}\right\|^2 &\leqslant \frac{E^2}{\bar n}\|\mu\|^2\left\|\sum\varphi_k\otimes\sqrt{\bar n+k}\mathrm{e}^{-\mathrm{i}\theta}P_{\bar n}\mathrm{e}^{\mathrm{i}k\theta}\right.\\ &\qquad\left.-\sum\varphi_k\otimes\sqrt{\bar n+k+1}\mathrm{e}^{\mathrm{i}\theta}P_{\bar n}\mathrm{e}^{\mathrm{i}k\theta}\right\|^2\\ &\leqslant 2\frac{E^2}{\bar n}\|\mu\|^2\left(\left\|\sum\varphi_k\otimes\sqrt{\bar n+k}P_{\bar n}\mathrm{e}^{\mathrm{i}k\theta}\right\|^2+\left\|\sum\varphi_k\otimes\sqrt{\bar n+k+1}P_{\bar n}\mathrm{e}^{\mathrm{i}k\theta}\right\|^2\right).\end{aligned} \tag{A9}$$

The vectors $\{\varphi_k\otimes\mathrm{e}^{\mathrm{i}k\theta}\}_k$ being mutually orthogonal in $\mathcal{K}$, we can use Pythagoras' theorem to obtain, denoting $c = 2E^2\|\mu\|^2$,

$$\begin{aligned}\left\|W_{\bar n}\sum\varphi_k\otimes\mathrm{e}^{\mathrm{i}k\theta}\right\|^2 &\leqslant \sum c\left(2+\frac{1}{\bar n}\right)\|\varphi_k\otimes P_{\bar n}\mathrm{e}^{\mathrm{i}k\theta}\|^2 + 2\frac{c}{\bar n}|k|\|\varphi_k\otimes P_{\bar n}\mathrm{e}^{\mathrm{i}k\theta}\|^2\\ &\leqslant \sum c\left(2+\frac{1}{\bar n}+\frac{c}{\omega^2}\right)\|\varphi_k\otimes\mathrm{e}^{\mathrm{i}k\theta}\|^2+\frac{\omega^2k^2}{\bar n^2}\|\varphi_k\otimes P_{\bar n}\mathrm{e}^{\mathrm{i}k\theta}\|^2\\ &= c\left(2+\frac{1}{\bar n}+\frac{c}{\omega^2}\right)\left\|\sum\varphi_k\otimes\mathrm{e}^{\mathrm{i}k\theta}\right\|^2+\frac{1}{\bar n^2}\left\|\left(-\mathbb{1}\otimes\mathrm{i}\omega\frac{\partial}{\partial\theta}P_{\bar n}\right)\sum\varphi_k\otimes\mathrm{e}^{\mathrm{i}k\theta}\right\|^2\end{aligned} \tag{A10}$$

where we have used that $2c|k|/\bar n\leqslant c^2/\omega^2+k^2\omega^2/\bar n^2$. Theorem A6 allows us to conclude that $H_{\bar n}$ is essentially self adjoint on $\mathcal{D}$. This completes the proof of theorem A1, as we have already shown that $H_{\bar n}\varphi\otimes\mathrm{e}^{\mathrm{i}k\theta}\longrightarrow K\varphi\otimes\mathrm{e}^{\mathrm{i}k\theta}$ for all $\varphi\in\mathcal{H}$ and $k\in\mathbb{Z}$ and as this convergence can be extended to the whole of $\mathcal{D}$ by linearity.

Appendix B. The $\bar n\to\infty$ limit of coherent states

In this appendix we prove that coherent states

$$|\alpha\rangle = \Phi_{\theta_0}^{(\bar n)}(\theta) = \mathrm{e}^{-|\alpha|^2/2}\sum_{n=0}^{\infty}\frac{|\alpha|^n}{\sqrt{n!}}\mathrm{e}^{\mathrm{i}(n-\bar n)(\theta-\theta_0)} \tag{B11}$$

with $\alpha = |\alpha|\mathrm{e}^{-\mathrm{i}\theta_0}$, are represented in Floquet theory, i.e. in the limit $|\alpha| = \sqrt{\bar n}\to\infty$, by a generalized function $\Phi_{\theta_0}(\theta)$, which is real, depends on $\theta-\theta_0$, and

$$(\Phi_{\theta_0}(\theta))^2 = 2\pi\delta(\theta-\theta_0). \tag{B12}$$

We prove this by showing that for any sufficiently smooth function $f(\theta)\in\mathcal{L} = L_2(\mathbb{S}^1,\mathrm{d}\theta/2\pi)$,

$$\lim_{|\alpha|\to\infty}\langle\alpha|f(\theta)|\alpha\rangle_{\mathcal{L}} = f(\theta_0) \tag{B13}$$

† Note that any vector in $\mathcal{D}$ can be written as such a sum.

and noting that for $\theta = \theta_0$ the coherent state (B11) is real. It is enough to prove that for any $k \in \mathbb{Z}$

$$\lim_{|\alpha|\to\infty} \langle\alpha|e^{ik\theta}|\alpha\rangle_{\mathcal{L}} = e^{ik\theta_0}. \tag{B14}$$

We can write

$$\begin{aligned}\langle\alpha|e^{ik\theta}|\alpha\rangle_{\mathcal{L}} &= e^{-|\alpha|^2}\sum_{n,n'=0}^{\infty}\frac{|\alpha|^{n+n'}}{\sqrt{n!n'!}}e^{-i(n-n')\theta_0}\int_0^{2\pi}\frac{d\theta}{2\pi}e^{i(n+k-n')\theta}\\ &= e^{ik\theta_0}e^{-|\alpha|^2}\sum_{n=0}^{\infty}\frac{|\alpha|^{2n+|k|}}{\sqrt{n!(n+|k|)!}}\end{aligned} \tag{B15}$$

where we have first exchanged the integral and the sum (since the sum is absolutely convergent), and then used

$$\int_0^{2\pi}\frac{d\theta}{2\pi}\exp(i(n+k-n')\theta) = \delta_{k,n'-n}.$$

The result follows thus from the

Lemma B1. For $k \geqslant 0$, the function

$$G(z) := e^{-z}\sum_{n=0}^{\infty}\frac{z^{n+k/2}}{\sqrt{n!(n+k)!}} \tag{B16}$$

satisfies

$$\lim_{z\to\infty} G(z) = 1. \tag{B17}$$

Proof. This result can be obtained by generalizing an argument used in [29]. We first remark that $\sqrt{n!(n+k)!} = n!\sqrt{(n+k)(n+k-1)\ldots(n+1)}$ and use for each of the factors the identity

$$\frac{1}{(n+j)^{s+1}} = \frac{1}{\Gamma(s+1)}\int_0^{\infty} dt\, t^s e^{-(n+j)t} \tag{B18}$$

(which is just the definition of the Γ-function) for $s = -1/2$. Using $\Gamma(1/2) = \pi^{1/2}$ we can thus write

$$((n+k)\ldots(n+1))^{-1/2} = \pi^{-k/2}\int_0^{\infty}dt_1\ldots\int_0^{\infty}dt_k\prod_{j=1}^{k}t_j^{-1/2}\exp\left(-\sum_{j=1}^{k}(n+j)t_j\right) \tag{B19}$$

and therefore

$$\begin{aligned}G(z) = \pi^{-k/2}z^{k/2}e^{-z}&\int_0^{\infty}dt_1\ldots\int_0^{\infty}dt_k\prod_{j=1}^{k}t_j^{-1/2}\exp\left(-\sum_{j=1}^{k}jt_j\right)\\ &\times\sum_{n=0}^{\infty}\frac{z^n}{n!}\left(\exp\left(-\sum_{j=1}^{k}t_j\right)\right)^n.\end{aligned} \tag{B20}$$

The last sum is equal to $\exp(z\exp(-\sum_{j=1}^{k}t_j))$.

Making the change of variables $\exp(-t_j) = 1 - x_j/z$, we obtain

$$G(z) = \pi^{-k/2} z^{k/2} \mathrm{e}^{-z} \int_0^z \mathrm{d}x_1 \ldots \int_0^z \mathrm{d}x_k \, z^{-k} \exp\left(z \prod_{j=1}^{k} (1 - x_j/z)\right)$$
$$\times \prod_{j=1}^{k} \left[\ln \frac{1}{1 - x_j/z}\right]^{-1/2} (1 - x_j/z)^{j-1}$$
$$= \pi^{-k/2} \int_0^\infty \mathrm{d}x_1 \ldots \int_0^\infty \mathrm{d}x_k \, \exp\left(-z\left(1 - \prod_{j=1}^{k}(1 - x_j/z)\right)\right)$$
$$\times \prod_{j=1}^{k} \chi_{[0;z]}(x_j) z^{-1/2} \left[\ln \frac{1}{1 - x_j/z}\right]^{-1/2} (1 - x_j/z)^{j-1}$$
$$= \pi^{-k/2} \int_0^\infty \mathrm{d}x_1 \ldots \int_0^\infty \mathrm{d}x_k \, f_z(x_1, \ldots, x_k) \tag{B21}$$

where $\chi_{[0;z]}(x)$ represents the characteristic function of the interval $[0; z]$. □

To prove lemma B1, we will use the theorem of dominated convergence of Lebesgue [50], a simplified version of which can be stated as follows.

Theorem B2. Let $\{f_n\}$ be a sequence of absolutely integrable functions over a subset X of $\mathbb{R}^k$ which converge pointwise for almost all points to a function f. Suppose that there exists a positive and integrable function g such that $|f(x)| \leqslant g(x)$ for almost every x. Then f is integrable and

$$\lim_{n\to\infty} \int_X |f_n - f| \mathrm{d}^k x = 0 \qquad \text{and} \qquad \lim_{n\to\infty} \int_X f_n \, \mathrm{d}^k x = \int_X f \, \mathrm{d}^k x. \tag{B22}$$

Note that instead of the limit $z \to \infty$ in (B17), we can consider this limit on any sequence tending to $+\infty$†.

Assuming that we have found a suitable function $g(x_1, \ldots, x_k)$ that bounds the integrand $f_z(x_1, \ldots, x_k)$ of $G(z)$, the argument proceeds simply as follows. First, note that we have

$$\lim_{z\to\infty} \exp\left(-z\left(1 - \prod_{j=1}^{k}(1 - x_j/z)\right)\right) = \exp\left(-\sum_{j=1}^{k} x_j\right). \tag{B23}$$

This can easily be seen by developing the product in the argument of the exponential. Next, for each $j \geqslant 1$, we have that

$$\lim_{z\to\infty} z^{-1/2} \left[\ln \frac{1}{1 - x_j/z}\right]^{-1/2} (1 - x_j/z)^{j-1} = x_j^{-1/2}. \tag{B24}$$

To obtain this result, we can take the square of this function and then use de l'Hospital's rule. Finally the limit of the characteristic function is simply the constant 1. Hence we have shown that

$$\lim_{z\to\infty} f_z(x_1, \ldots, x_k) = \prod_{j=1}^{\infty} x_j^{-1/2} \exp(-x_j) \qquad \text{on } (0; \infty). \tag{B25}$$

Therefore, by theorem B2, we have

$$\lim_{z\to\infty} G(z) = \pi^{-k/2} \prod_{j=1}^{k} \int_0^\infty \mathrm{d}x_j \, x_j^{-1/2} \exp(-x_j) = \pi^{-k/2} \Gamma(1/2)^k = 1. \tag{B26}$$

† In fact, as it can readily be seen, the arguments below are actually independent of the choosen sequence.

To conclude the proof, it remains to show the existence of the bounding function g. We will bound each factor in f_z separately. First, notice that f_z is a positive function. Next, we have

$$(1 - x_j/z)^{j-1}\chi_{[0;z]}(x_j) \leqslant 1. \tag{B27}$$

Using the inequality $y \leqslant -\ln(1-y)$ which is valid in $0 \leqslant y < 1$, we find that

$$z^{-1/2}\left[\ln \frac{1}{1 - x_j/z}\right]^{-1/2} \leqslant x_j^{-1/2} \qquad \text{for } 0 < x_j \leqslant z. \tag{B28}$$

Finally, we have in the interval $0 \leqslant x_j \leqslant z$,

$$\exp\left(-z\left(1 - \prod_{j=1}^{k}(1 - x_j/z)\right)\right) \leqslant \exp\left(-\frac{1}{k}\sum_{j=1}^{k} x_j\right). \tag{B29}$$

Indeed, taking the logarithm and denoting $y_j = x_j/z$, we see that this inequality is equivalent to

$$1 - \prod_{j=1}^{k}(1 - y_j) \geqslant \frac{1}{k}\sum_{j=1}^{k} y_j \qquad \text{for } 0 \leqslant y_j \leqslant 1. \tag{B30}$$

This inequality follows directly from the fact that (identifying $1 - y_j$ with s_j)

$$\prod_{j=1}^{k} s_j \leqslant \left(\prod_{j=1}^{k} s_j\right)^{\frac{1}{k}} \leqslant \frac{1}{k}\sum_{j=1}^{k} s_j \qquad \text{for } 0 \leqslant s_j \leqslant 1. \tag{B31}$$

The first inequality is true because each s_j belongs to the interval [0; 1], and the second follows from the fact that a geometric mean is always less than or equal to an arithmetic mean†. Thus, we have shown that

$$0 \leqslant f_z(x_1, \ldots, x_k) \leqslant \exp\left(-\frac{1}{k}\sum_{j=1}^{k} x_j\right)\prod_{j=1}^{\infty} x_j^{-1/2} =: g(x_1, \ldots, x_k). \tag{B32}$$

Now, the bounding function g is integrable, since

$$\int_0^\infty \mathrm{d}x_1 \ldots \int_0^\infty \mathrm{d}x_k\, g(x_1, \ldots, x_k) = \left(\int_0^\infty x^{-1/2}\mathrm{e}^{-x/k}\,\mathrm{d}x\right)^k = (\pi k)^{k/2} < \infty \tag{B33}$$

which completes the proof.

Appendix C. The formal calculus of $\boldsymbol{\delta_{1/2}(\theta - \theta_0)}$

Since the origin of the $\delta_{1/2}$ function is

$$\begin{aligned} |\Phi_{\theta_0}(\theta)| &\underset{\bar{n}\to\infty}{\rightsquigarrow} (8\pi)^{1/4}\bar{n}^{1/4}\exp\{-2\bar{n}[1 - \cos(\theta - \theta_0)]\} \\ &\underset{\bar{n}\to\infty}{\rightsquigarrow} (2\pi)^{1/2}\frac{(2\bar{n})^{1/4}}{\pi^{1/4}}\mathrm{e}^{-\bar{n}(\theta-\theta_0)^2} \\ &\xrightarrow[\bar{n}\to\infty]{} (2\pi)^{1/2}\delta_{1/2}(\theta - \theta_0) \end{aligned} \tag{C34}$$

we can establish the following formal calculus rules:

(1)

$$(\delta_{1/2}(\theta - \theta_0))^2 = \delta(\theta - \theta_0). \tag{C35}$$

† This can easily be shown by using the concavity of the function $\ln(x)$.

(2) If $\theta \neq \theta_0$

$$\delta_{1/2}(\theta - \theta_0) = 0. \tag{C36}$$

(3) For any $\xi \in \mathcal{L}$

$$\delta_{1/2}(\theta - \theta_0)\xi(\theta) = \delta_{1/2}(\theta - \theta_0)\xi(\theta_0). \tag{C37}$$

(4)

$$2\pi \langle \delta_{1/2}(\theta - \theta_0) | \delta_{1/2}(\theta - \theta_0) \rangle_{\mathcal{L}} = 1. \tag{C38}$$

(5) For any $\xi \in \mathcal{L}$

$$\langle \delta_{1/2}(\theta - \theta_0) | \xi(\theta) \rangle_{\mathcal{L}} = 0. \tag{C39}$$

(6) For any observable $F : \mathcal{L} \to \mathcal{L}$ that is a continuous function of θ,

$$2\pi \langle \delta_{1/2}(\theta - \theta_0) | F(\theta) | \delta_{1/2}(\theta - \theta_0) \rangle_{\mathcal{L}} = F(\theta_0). \tag{C40}$$

(7) By equation (39), we obtain

$$\left\langle \delta_{1/2}(\theta - \theta_0) \left| -\mathrm{i}\frac{\partial}{\partial \theta} \right| \delta_{1/2}(\theta - \theta_0) \right\rangle_{\mathcal{L}} = 0. \tag{C41}$$

(8) If $\varphi \in \mathcal{H}$ and $M : \mathcal{K} \to \mathcal{K}$ is an observable that with respect to θ is a multiplication operator, continuous in θ, then taking for θ a particular value θ_0 defines a family of operators $M(\theta_0) : \mathcal{H} \to \mathcal{H}$, parametrized by θ_0. Then

$$2\pi \langle \varphi \otimes \delta_{1/2}(\theta - \theta_0) | M(\theta) | \varphi \otimes \delta_{1/2}(\theta - \theta_0) \rangle_{\mathcal{K}} = \langle \varphi | M(\theta_0) | \varphi \rangle_{\mathcal{H}}. \tag{C42}$$

We remark that $\delta_{1/2}(\theta - \theta_0)$ can be interpreted as a linear functional on the space of continuous observables $F = F^\dagger : \mathcal{L} \to \mathcal{L}$ by

$$\delta_{1/2,\theta_0} : F \mapsto \langle \delta_{1/2}(\theta - \theta_0) | F(\theta) | \delta_{1/2}(\theta - \theta_0) \rangle_{\mathcal{L}}. \tag{C43}$$

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