Floquet theory

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1 Introduction

2 The Floquet formalism: quasienergies and quasi-stationary states

In this section we outline the basic formalism of Floquet physics, following the notation in [1]. As is mentioned in the introduction, Floquet effects happen with a time-periodic Hamiltonian; below we let $T = 2\pi/\omega$ be the period. Such a Hamiltonian is usually an effective Hamiltonian when the system (hereafter "matter") is coupled with another degree of freedom which does not change much in the time evolution; the latter is hereafter called "light", since in condensed matter systems, periodic driving is usually achieved by shedding a beam of light to the matter: consider the general form of light-matter interaction Hamiltonian with only one active photon mode

$$H_{\text{full}} = H \otimes 1_{\text{light}} + 1_{\text{matter}} \otimes \hbar\omega \left(b^\dagger b + \frac{1}{2} \right) + \underbrace{bV + b^\dagger V^\dagger}_{H_{\text{light-matter coupling}}}, \tag{1}$$

and we assume that the state of the electromagnetic part is close to a coherent state $|\alpha e^{-i\omega t}\rangle$ with strong intensity that almost has zero time evolution. Under this assumption, we can project out the electromagnetic degree of freedom by

$$P = \sum_i |i\rangle \langle i| \otimes \langle \alpha e^{-i\omega t}| , \tag{2}$$

where i labels eigenstates of the matter degrees of freedom, and this means the Hamiltonian for the matter part is (TODO: the additional term $-i\hbar U \partial_t U^\dagger$)

$$H_{\text{driven}} = P H_{\text{full}} P^\dagger = H + \underbrace{\hbar\omega|\alpha|^2}_{\text{const.}} + \alpha V e^{-i\omega t} + \alpha^* V^\dagger e^{i\omega t}, \tag{3}$$

which indeed evolves with period $2\pi/\omega$.

From the Floquet theory of differential equation, we know it is possible to expand an arbitrary state that evolves according to H into a linear combination (the coefficients are constants) of $\{ |\psi_n(t)\rangle \}$ where

$$|\psi_n(t)\rangle = e^{-i\varepsilon_n t/\hbar} |\Phi_n(t)\rangle , \quad |\Phi_n(t+T)\rangle = |\Phi_n(t)\rangle . \tag{4}$$

By discrete periodicity of $|\Phi_n(t)\rangle$ we make Fourier expansion

$$|\Phi_n(t)\rangle = \sum_m e^{-im\omega t} |\phi_n^{(m)}\rangle , \tag{5}$$

where m goes over all integers. Note that here $|\phi_n^{(m)}\rangle$ are *Fourier coefficients* and are not eigenstates of anything; there is no normalization or orthogonality condition for them. Using i to label the eigenstates of the matter, we have

$$|\Phi_n(t)\rangle = \sum_i \sum_m e^{-im\omega t} \langle i|\phi_n^{(m)}\rangle \, |i\rangle . \tag{6}$$

The coefficients before $|i\rangle$, not coefficients before $|\phi_n^{(m)}\rangle$ in (5), give the expansion of $|\Phi\rangle$ in a complete, orthogonal basis. The significance of $|\phi\rangle$ vectors can be seen immediately below.

The Schrodinger equation

$$\frac{d}{dt} |\psi_n(t)\rangle = H |\psi_n(t)\rangle \tag{7}$$

now reads

$$(\varepsilon_n + m\hbar\omega) |\phi_n^{(m)}\rangle = \sum_{m'} H^{(m-m')} |\phi_n^{(m')}\rangle , \tag{8}$$

where

$$H(t) = \sum_m \mathrm{e}^{-\mathrm{i}m\omega t} H^{(m)}. \tag{9}$$

Thus we find

$$\varepsilon_n |\phi_n^{(m)}\rangle = \sum_{m'} (H^{(m-m')} - m\hbar\omega\delta_{mm'}) |\phi_n^{(m')}\rangle . \tag{10}$$

Note that this equation is an eigenvalue problem in the *extended Hilbert space*: the component $\langle i|\phi_n^{(m)}\rangle$ is labeled by both m and i. The eigenvalues ε_n are known as the *Floquet quasienergy* of the *Floquet quasi-stationary state (or quasi-eigenstate)* $|\Psi_n(t)\rangle$, which can be obtained by diagonalizing

$$H_{\text{Floquet},mm'} = H^{(m-m')} - m\hbar\omega\delta_{mm'}. \tag{11}$$

(11) looks like a light-matter interaction Hamiltonian written in operator form for the matter part and in the Fock basis for the light part (labeled by m and m', which look like photon numbers); when a cutoff on m is applied, it appears to be an effective Hamiltonian where photon degrees of freedom have been integrated out. However, unlike conventional effective Hamiltonians whose eigenstates can in principle be obtained by applying a projection operator on a subset of eigenstates of the full Hamiltonian, eigenstates of (11) *do not* correspond to any eigenstate of the full Hamiltonian e.g. (1): the light part is in a coherent state, which is far from any eigenstate of the linear electromagnetic Hamiltonian $\hbar\omega(n+\frac{1}{2})$. Instead, Floquet formalisms is to be understood in a more generic framework of non-equilibrium physics: Floquet Green function can be calculated within the Keldysh formalism, and (11) can be understood as the non-equilibrium self-energy [2, 3] and therefore is not necessarily an equilibrium effective Hamiltonian.

Finally, we go on to characterize the structure of the solutions of (10). The difference between the Floquet effective Hamiltonian (11) and conventional, "equilibrium" effective Hamiltonians can also be seen by the structure of its eigenstates, because the dimension of (11), fully expanded into its matrix elements, is the number of the values of m considered times the dimension of the matter Hilbert space, and thus (11)'s eigenstates are overcomplete. We can actually point out where overcompletion appears: note that if ε_n satisfies (4), then so does $\varepsilon_n + m\hbar\omega$. We can directly prove that if $(\varepsilon, \{\,|\phi^{(m)}\rangle\}_m)$ is a solution of (10), then so is $(\varepsilon + \hbar m'\omega, \{\,|\phi^{(m+m')}\rangle\}_m)$. This fact can also be proved by noticing that

$$|\psi_n(t)\rangle = \mathrm{e}^{-\mathrm{i}\varepsilon_n t/\hbar} \sum_m \mathrm{e}^{-\mathrm{i}m\omega t} |\phi_n^{(m)}\rangle = \mathrm{e}^{-\mathrm{i}(\varepsilon+\hbar m'\omega)t/\hbar} \sum_m \mathrm{e}^{-\mathrm{i}m\omega t} |\phi_n^{(m+m')}\rangle . \tag{12}$$

Therefore, the spectrum of (11) has redundancy. There however is no redundant information in $\{\,|\phi_n^{(m)}\rangle\}$, because $\langle i|\phi_n^{(m)}\rangle$ is the $m\omega$-frequency component of $|\Phi_n(t)\rangle$ projected on the basis vector $|i\rangle$, and every $|\phi_n^{(m)}\rangle$ is needed to determine $|\Phi_n(t)\rangle$ and thus $|\psi_n(t)\rangle$.

Although there is no generic orthogonal relation pertaining to $\{\,|\psi_n(t)\rangle\}$, after time averaging an orthogonal relation can be obtained. From the fact that (11) is Hermitian, we have

$$\sum_m \langle \phi_n^{(m)}|\psi_{n'}^{(m)}\rangle = \delta_{nn'}, \tag{13}$$

and therefore the time average of $\langle\psi_n(t)|\psi_{n'}(t)\rangle$ is

$$\begin{aligned}
\overline{\langle\psi_n(t)|\psi_{n'}(t)\rangle} &= \lim_{T\to\infty} \frac{1}{T}\int_0^T \mathrm{d}t\, \mathrm{e}^{\mathrm{i}(\varepsilon_n-\varepsilon_{n'})t} \sum_{m,m'} \mathrm{e}^{\mathrm{i}(m-m')t} \langle\phi_n^{(m)}|\phi_{n'}^{(m')}\rangle \\
&= \sum_{m,m'} \delta_{\varepsilon_n,\varepsilon_{n'}} \delta_{mm'} \langle\phi_n^{(m)}|\phi_{n'}^{(m')}\rangle \\
&= \delta_{\varepsilon_n,\varepsilon_{n'}} \sum_m \langle\phi_n^{(m)}|\psi_{n'}^{(m)}\rangle = \delta_{nn'}.
\end{aligned} \tag{14}$$

Therefore, after time averaging, the Floquet quasi-stationary states are orthogonal to each other.

In conclusion, a Floquet system is an inherently non-equilibrium system, but a set of quasi-eigenstates $\{|\psi_n\rangle\}$ with orthogonality relation (14) can still be well defined, the number of which is the same as the dimension of the Hilbert space (*not* the extended Hilbert space). The quasi-eigenstates and their quasi-energies can be found by solving the eigenvalue problem (10) in the extended Hilbert space (*not* the Hilbert space) and then putting the resulting $(\varepsilon_n, \{\phi_n^{(m)}\})$ into (4). For each quasi-eigenstate, we have countable infinite quasi-energies, the difference between the nearest two being $\hbar\omega$; thus all distinct Floquet quasi-eigenstates can be indexed by quasi-energies that are within one "Floquet-Brillouin zone".

3 Floquet formalisms compared with conventional treatment of periodic driving

Periodic driving can also be construed in alternative approaches. In this section we review two of them and discuss the advantage of explicitly introducing Floquet quasienergies and quasi-stationary states. First, periodic driving is in principle well captured by time-dependent perturbation theory. For example, for a two-level system driven by $E = E_0 \cos\omega t$,

$$H = \omega_{\mathrm{eg}} |\mathrm{e}\rangle\langle\mathrm{e}| - \Omega |\mathrm{e}\rangle\langle\mathrm{g}| \,\mathrm{e}^{-\mathrm{i}\omega t} + \text{h.c.}, \quad \Omega = -\frac{\mu_{\mathrm{eg}} E_0}{2\hbar}, \tag{15}$$

the linear response of the dipole $\boldsymbol{\mu} = \mu_{\mathrm{eg}} |\mathrm{e}\rangle\langle\mathrm{g}| + \text{h.c.}$ to the driving field can be straightforwardly calculated in time-dependent perturbation theory; the final result is (for convenience we assume that μ_{eg} and Ω are all real)

$$\langle\mu\rangle^{(1)}(t) = \mu_{\mathrm{eg}}\Omega \left(\frac{\mathrm{e}^{\mathrm{i}\omega t}}{\omega_{\mathrm{eg}} + \omega - \mathrm{i}\Gamma_{\mathrm{e}}} + \frac{\mathrm{e}^{-\mathrm{i}\omega t}}{\omega_{\mathrm{eg}} - \omega - \mathrm{i}\Gamma_{\mathrm{e}}} + \frac{\mathrm{e}^{-\mathrm{i}\omega t}}{\omega_{\mathrm{eg}} + \omega + \mathrm{i}\Gamma_{\mathrm{e}}} + \frac{\mathrm{e}^{\mathrm{i}\omega t}}{\omega_{\mathrm{eg}} - \omega + \mathrm{i}\Gamma_{\mathrm{e}}}. \right) \tag{16}$$

The Hamiltonian (15) can also be understood in the framework of Floquet formalism: $H^{(0)}$ is just $\omega_{\mathrm{eg}} |\mathrm{e}\rangle\langle\mathrm{e}|$, and $H^{(1)}$ is $\Omega |\mathrm{e}\rangle\langle\mathrm{g}|$. Our goal is to understand how the two formalisms are equivalent to each other. At the order of (16), where only the $\mathrm{e}^{\pm\omega t}$ components are considered, we can restrict ourselves on the following sub-matrix of the full Floquet Hamiltonian (from left to right we display g and e components with $m = -1, 0, 1$):

$$\begin{pmatrix} \omega & 0 & 0 & -\Omega & 0 & 0 \\ 0 & \omega_{\mathrm{eg}} + \omega & -\Omega & 0 & 0 & 0 \\ 0 & -\Omega & 0 & 0 & 0 & -\Omega \\ -\Omega & 0 & 0 & \omega_{\mathrm{eg}} & -\Omega & 0 \\ 0 & 0 & 0 & -\Omega & -\omega & 0 \\ 0 & 0 & -\Omega & 0 & 0 & \omega_{\mathrm{eg}} - \omega \end{pmatrix}, \tag{17}$$

and moreover, since only linear dependence on Ω is included in (16), we also only consider the first-order correction of $|\mathrm{e}\rangle$ and $|\mathrm{g}\rangle$ (recall that the possible number of Floquet quasi-stationary states is the same as the dimension of the Hilbert space, and in this case, is two). Below we use $|\tilde{\mathrm{e}}\rangle$, $|\tilde{\mathrm{g}}\rangle$ to refer to the Floquet-corrected states. The $m = -$ part of $|\tilde{\mathrm{g}}\rangle$, because of the position of the Ω matrix elements, is

$$|\phi_{\mathrm{g}}^{(-1)}\rangle = \frac{-\Omega}{\omega - \omega_{\mathrm{eg}}} |\mathrm{e}\rangle\,, \tag{18}$$

and similarly we have

$$|\phi_{\mathrm{g}}^{(1)}\rangle = \frac{-\Omega}{-\omega - \omega_{\mathrm{eg}}} |\mathrm{e}\rangle\,. \tag{19}$$

Assuming that $|\tilde{\mathrm{g}}\rangle$ is the occupied state, the corresponding value of $\langle\mu\rangle$ is

$$\begin{aligned} \langle\mu\rangle^{(1)}(t) &= \langle\tilde{\mathrm{g}}(t)|\mu|\tilde{\mathrm{g}}(t)\rangle = \langle\mathrm{g}|\mu|\phi_{\mathrm{g}}^{(1)}\rangle\,\mathrm{e}^{-\mathrm{i}\omega t} + \langle\mathrm{g}|\mu|\phi_{\mathrm{g}}^{(-1)}\rangle\,\mathrm{e}^{\mathrm{i}\omega t} + \text{h.c.} \\ &= \mu_{\mathrm{eg}}\Omega \left(\frac{\Omega}{\omega_{\mathrm{eg}} - \omega} \mathrm{e}^{-\mathrm{i}\omega t} + \frac{\Omega}{\omega_{\mathrm{eg}} + \omega} \mathrm{e}^{\mathrm{i}\omega t} + \text{h.c.} \right). \end{aligned} \tag{20}$$

Ignoring the imaginary parts, this is exactly the same as (16). Therefore, Floquet formalism is beneficial when the period external driving field is strong, and the conventional concept of perturbation expansion of the system's response to the driving field is no longer practically feasible.

The rotational wave approximation (RWA) is also used when a periodic pumping is present; here we show that it can be seen as a degeneracy case of the Floquet formalism.

4 Floquet effects in spectroscopy

Ordinary angle-resolved photoemission spectroscopy (ARPES) spectra

$$I(\mathbf{k}, \omega) \propto \sum_{\mathbf{k}', c} \left|M_{\mathbf{k}\mathbf{k}'}^{fc}\right|^2 \delta(\omega - \epsilon_{c\mathbf{k}'}) f(\omega), \tag{21}$$

where $(f, \mathbf{k})$ is the out-coming electron mode, $(c, \mathbf{k}')$ is an electronic mode within the material and $f(\omega)$ is the Fermi-Dirac distribution, are usually calculated by Fermi golden rule [4]; when the starting state is non-equilibrium, the spectral function should be replaced by the lesser Green function and we get [5, 6, 7] TODO: cross term where $c \neq c'$

$$I(t, \mathbf{k}, \omega) \propto -\mathrm{i} \int_{t_0}^{t} \mathrm{d}t_1 \int_{t_0}^{t} \mathrm{d}t_2 \, s(t_1) s(t_2) \left|M_{\mathbf{k}\mathbf{k}'}^{fc}\right|^2 G_{\mathbf{k}'}^{<}(t_2, t_1) \mathrm{e}^{\mathrm{i}\omega(t_1 - t_2)}, \tag{22}$$

where $s(t)$ is the envelope of the probe pulse. When the system indeed stays in equilibrium, and the probe pulse is long enough and hence $s(t)$ can be seen as a constant and $t \to \infty$, it can be directly verified that (22) reduces to (21). When the system is Floquet-driven, (21) is not correct, but in the long probe pulse limit, (22) becomes

$$I(t \to \infty, \mathbf{k}, \omega) \propto -\mathrm{i} \int \mathrm{d}\bar{t} \int \mathrm{d}\tau \, G_{\mathbf{k}'}^{<}(\bar{t}, \bar{t} + \tau) \mathrm{e}^{\mathrm{i}\omega\tau} = -\mathrm{i} \left\langle \int \mathrm{d}\tau \, \mathrm{e}^{\mathrm{i}\omega\tau} G_{\mathbf{k}'}^{<}(\bar{t}, \bar{t} + \tau) \right\rangle_{\bar{t}}, \tag{23}$$

or in other words, the time-averaged occupied density of states. This, then, can be determined by the Floquet quasi-stationary states and quasienergies. We have [1]

$$\bar{\rho}(\Omega) = -\mathrm{i} \left\langle \int \mathrm{d}\tau \, \mathrm{e}^{\mathrm{i}\omega\tau} G_{\mathbf{k}}^{<} \right\rangle = \sum_{n\mathbf{k}, m} A_{n\mathbf{k}}^{(m)} \delta\left(\varepsilon_{n\mathbf{k}} + m\omega - \Omega\right), \quad A_{n\mathbf{k}}^{(m)} = \langle \phi_{n\mathbf{k}}^{(m)} | \phi_{n\mathbf{k}}^{(m)} \rangle. \tag{24}$$

The ARPES spectrum of a Floquet-driven band structure, therefore, is given by all Floquet bands weighted by the magnitude of $|\phi_{n\mathbf{k}}^{(m)}\rangle$.

TODO: But then what about non-diagonal terms in Floquet Hamiltonian? Note that $n \neq n'$ are not considered above.

Interestingly, it is possible to use light to stimulate some long-lived degrees of freedom in a solid and let it drive the rest of the system, which sometimes is known as "self-driving". In [7], it is demonstrated that Floquet renormalization of the band structure can be observed even long after the pump is turned off, which is due to the existence of excitons previously created by the pump. TODO: exciton captured by electron self-energy; exciton creates Σ_{cv}

In principle there are indefinite exciton-driven Floquet quasi-band ARPES signatures, but since the signatures are weighted by $A_{n\mathbf{k}}^{(m)}$, which are usually small when $|m|$ is large, usually only the $m = 1$ Floquet signatures are important, which can be calculated from Fermi golden rule in a more conventional way.

TODO: can band renormalization be captured by Fermi golden rule?

References

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