Floquet physics

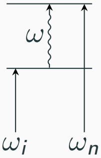
Periodic driving, formalism, and spetroscopy

Jinyuan Wu

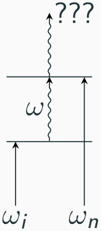
December 11, 2023

Introduction

Time-dependent perturbation theory, $\cancel{\omega_{eg} + \omega}$
$\Rightarrow$ Fermi golden rule (finite wave packet or not)



What happens when we consider high order perturbations?



Inherently non-equilibrium The state of photons is a coherent state: $|\Psi\rangle$ far from any eigenstate!

Overview

- From Schrodinger equation to Floquet effective Hamiltonian
- Relation with time-dependent perturbation theory and rotating wave approximation (RWA)?
- Floquet correction to electron band

The Floquet formalism

Quasi-stationary states and quasienergies

Periodically driven Hamiltonian: quasi-eigensystem

Floquet theory $H(t) = H(t+T) \Rightarrow$ for every $|\psi(t)\rangle$,

$$|\psi(t)\rangle = \sum_n |\psi_n(t)\rangle\,, \quad |\psi_n(t)\rangle = \mathrm{e}^{-\mathrm{i}\,\varepsilon_n t/\hbar}\,\underbrace{|\Phi_n(t)\rangle}_{\text{period } T},$$

$$|\Phi_n(t)\rangle = \underbrace{\sum_m \mathrm{e}^{-\mathrm{i}\,m\omega t}\,|\phi_n^{(m)}\rangle}_{\text{discrete Fourier series}}, \quad \omega = 2\pi/T.$$

Highlights

- $\mathcal{H} \otimes \{m = \ldots, -1, 0, 1, \ldots\}$: **extended Hilbert space**
- $\{\Phi_n(t)\}$: **quasi-stationary states**
- $\{\varepsilon_n\}$: **quasienergies**

Our task How to $\{\phi_n^{(m)}\}$ and $\{\varepsilon_n\}$?

Floquet effective Hamiltonian

Our task A Hamiltonian for $\{\phi_n^{(m)}\}$ and $\{\varepsilon_n\}$?

$$\underbrace{H(t)}_{=:\sum_m \mathrm{e}^{-\mathrm{i}\, m\omega t} H^{(m)}} \qquad |\psi_n(t)\rangle = \mathrm{i}\,\hbar\partial_t\,|\psi_n(t)\rangle = \varepsilon_n + \mathrm{i}\,\hbar\partial_t\,|\Phi_n(t)\rangle$$

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

Floquet effective Hamiltonian Indeed we have a Hamiltonian!!

Floquet effective Hamiltonian

Our task A Hamiltonian for $\{\phi_n^{(m)}\}$ and $\{\varepsilon_n\}$?

$$\underbrace{H(t)}_{=:\sum_m \mathrm{e}^{-\mathrm{i}\, m\omega t} H^{(m)}} \qquad |\psi_n(t)\rangle = \mathrm{i}\,\hbar\partial_t\,|\psi_n(t)\rangle = \varepsilon_n + \mathrm{i}\,\hbar\partial_t\,|\Phi_n(t)\rangle$$

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

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

Floquet effective Hamiltonian Indeed we have a Hamiltonian!!

Floquet effective Hamiltonian

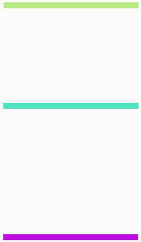
ε_n, $(\cdots, |\phi_n^{(-1)}\rangle, |\phi_n^{(0)}\rangle, |\phi_n^{(1)}\rangle, \cdots)$ are obtained by diagonalizing

$$
\begin{array}{c}
 \\ m=-2 \\ m=-1 \\ m=0 \\ m=1 \\ \\
\end{array}
\begin{array}{c}
\begin{array}{cccc} m'=-2 & m'=-1 & m'=0 & m'=1 \end{array} \\
\begin{pmatrix}
 & & \vdots & \vdots & & \\
 & H^{(0)}+2\hbar\omega & H^{(-1)} & H^{(-2)} & H^{(-3)} & \\
\dots & H^{(1)} & H^{(0)}+\hbar\omega & H^{(-1)} & H^{(-2)} & \dots \\
\dots & H^{(2)} & H^{(1)} & H^{(0)} & H^{(-1)} & \dots \\
 & H^{(3)} & H^{(2)} & H^{(1)} & H^{(0)}-\hbar\omega & \\
 & & \vdots & \vdots & &
\end{pmatrix}
\end{array}
$$

- Each "element" is a Hamiltonian on $\mathcal{H}$
- The hole H^{Floquet} is on the extended Hilbert space

Floquet Brillouin zone

Redundancy in $H^{\textbf{Floquet}}$ The number of independent quasienergies is not really multiplied by Floquet subspaces.



$$\mathrm{e}^{-\mathrm{i}(\varepsilon_n - m\hbar\omega)t}\,\underbrace{\mathrm{e}^{\mathrm{i}\,m\hbar\omega t}\,|\Phi_n(t)\rangle}_{\text{still periodic!}}$$

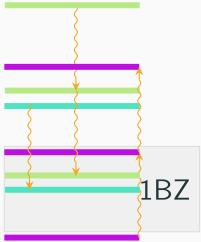
The number of independent quasi-stationary states $= \dim \mathcal{H}$

So only *one* energy Brillouin zone is needed.

But all $\phi_n^{(m)}$ are all needed to decide $|\Phi_n(t)\rangle$.

Floquet Brillouin zone

Redundancy in $H^{\textbf{Floquet}}$ The number of independent quasienergies is not really multiplied by Floquet subspaces.



$$e^{-\mathrm{i}(\varepsilon_n - m\hbar\omega)t} \underbrace{e^{\mathrm{i}\, m\hbar\omega t} \, |\Phi_n(t)\rangle}_{\text{still periodic!}}$$

The number of independent quasi-stationary states $= \dim \mathcal{H}$

So only *one* energy Brillouin zone is needed.

But all $\phi_n^{(m)}$ are all needed to decide $|\Phi_n(t)\rangle$.

Floquet formalism in eyes of other formalisms

“full” Floquet theory, perturbation theory, and RWA

Floquet formalism in hierarchy of approximations

Other ways to describe periodic driving

- Time-dependent perturbation theory (TD-PT)
- Rotating wave approximation (RWA)

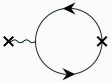
$$\begin{tikzcd}
\text{Keldysh, etc.} \arrow[r, "T=0"] & \text{TD-PT} \arrow[r, "\text{periodic, no cutoff}"] \arrow[dl] \arrow[d] \arrow[rrr, bend right] & \text{Floquet} \arrow[r, "\text{small }\Omega,\Delta"] & \text{RWA} \\
 & \text{various cutoff schemes} & &
\end{tikzcd}$$

Floquet formalism v.s. TD-PT

Response from time-dependent perturbation theory = response from $T = 0$ Feynman diagrams.

Example: first-order PT = Lindhard response function

$$\langle \mu^{(1)} \rangle = \mu_{\mathrm{eg}} \underbrace{\Omega}_{\text{Rabi freq.}} \left(\frac{\Omega}{\omega_{\mathrm{eg}} - \omega} \mathrm{e}^{-\mathrm{i}\omega t} + \frac{\Omega}{\omega_{\mathrm{eg}} + \omega} \mathrm{e}^{\mathrm{i}\omega t} + \mathrm{h.c.} \right) =$$

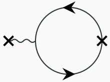


Floquet formalism v.s. TD-PT

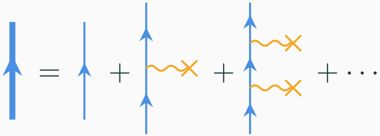
Response from time-dependent perturbation theory = response from $T = 0$ Feynman diagrams.

Example: first-order PT = Lindhard response function

$$\langle \mu^{(1)} \rangle = \mu_{\mathrm{eg}} \underbrace{\Omega}_{\text{Rabi freq.}} \left(\frac{\Omega}{\omega_{\mathrm{eg}} - \omega} \mathrm{e}^{-\mathrm{i}\omega t} + \frac{\Omega}{\omega_{\mathrm{eg}} + \omega} \mathrm{e}^{\mathrm{i}\omega t} + \mathrm{h.c.} \right) =$$



And when we sum over all of them...

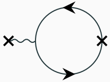


Floquet formalism v.s. TD-PT

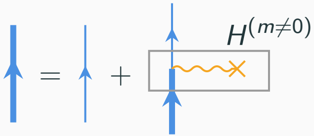
Response from time-dependent perturbation theory = response from $T = 0$ Feynman diagrams.

Example: first-order PT = Lindhard response function

$$\langle \mu^{(1)} \rangle = \mu_{\mathrm{eg}} \underbrace{\Omega}_{\text{Rabi freq.}} \left(\frac{\Omega}{\omega_{\mathrm{eg}} - \omega} \mathrm{e}^{-\mathrm{i}\omega t} + \frac{\Omega}{\omega_{\mathrm{eg}} + \omega} \mathrm{e}^{\mathrm{i}\omega t} + \mathrm{h.c.} \right) =$$



$H^{\textbf{Floquet}}$ **is the non-equilibrium self-energy**

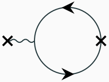


Floquet formalism v.s. TD-PT

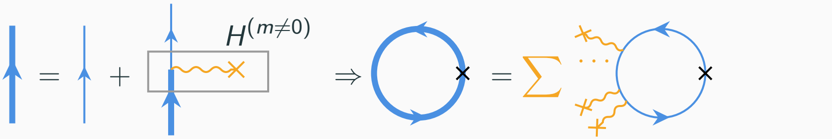
Response from time-dependent perturbation theory = response from $T = 0$ Feynman diagrams.

Example: first-order PT = Lindhard response function

$$\langle \mu^{(1)} \rangle = \mu_{\text{eg}} \underbrace{\Omega}_{\text{Rabi freq.}} \left(\frac{\Omega}{\omega_{\text{eg}} - \omega} \mathrm{e}^{-\mathrm{i}\omega t} + \frac{\Omega}{\omega_{\text{eg}} + \omega} \mathrm{e}^{\mathrm{i}\omega t} + \text{h.c.} \right) =$$



H^{Floquet} is the non-equilibrium self-energy



In the full H^{Floquet}, automatically all PT terms are considered!

Floquet formalism v.s. RWA

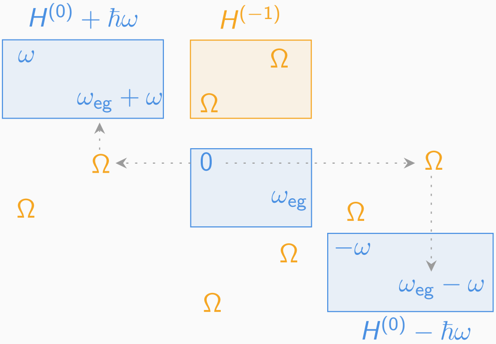
$H^{(0)} + \hbar\omega$ $\quad$ $H^{(-1)}$

$$\begin{matrix} \omega & & & \Omega & & \\ & \omega_{\text{eg}} + \omega & \Omega & & & \\ & \Omega & 0 & & & \Omega \\ \Omega & & & \omega_{\text{eg}} & \Omega & \\ & & & \Omega & -\omega & \\ & & \Omega & & & \omega_{\text{eg}} - \omega \end{matrix}$$

$H^{(0)} - \hbar\omega$

From Floquet to RWA

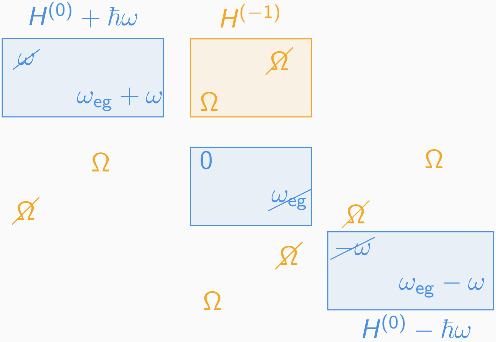
Floquet formalism v.s. RWA



From Floquet to RWA

- Small coupling: only first-order transitions from $|g\rangle$ matters

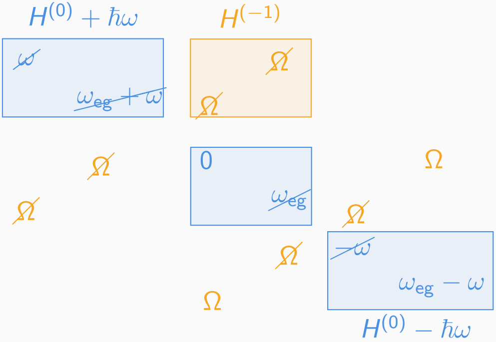
Floquet formalism v.s. RWA



From Floquet to RWA

- Small coupling: only first-order transitions from $|g\rangle$ matters

Floquet formalism v.s. RWA

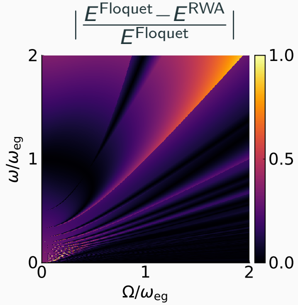


From Floquet to RWA

- Small coupling: only first-order transitions from $|g\rangle$ matters
- Near resonance: only the $m = 1$ $\omega_{eg} - \omega$ state matters

Floquet formalism v.s. RWA

$$H^{\mathrm{RWA}} = \begin{pmatrix} 0 & \Omega \\ \Omega & \omega_{\mathrm{eg}} - \omega \end{pmatrix}.$$



From Floquet to RWA

- Small coupling: only first-order transitions from $|\mathrm{g}\rangle$ matters
- Near resonance: only the $m = 1$ $\omega_{\mathrm{eg}} - \omega$ state matters

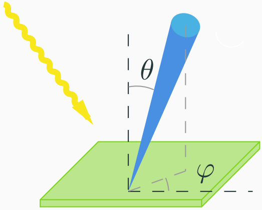
Indeed when $\Omega/\omega_{\mathrm{eg}} \lesssim 0.5$, $\omega/\omega_{\mathrm{eg}} \sim 1$, RWA works the best!

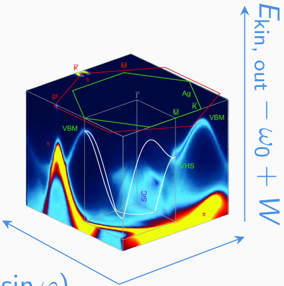
ARPES of Floquet systems

Are Floquet states "real"?

Angle-resolved photoemission spectroscopy (ARPES)

ARPES sheds *probe* beam to material (possibly *pumped* by another beam) and detects output electrons' $(\mathbf{k}, E)$[a]





$$\mathbf{k}_{\parallel} = k(\sin\theta\cos\varphi, \sin\theta\sin\varphi)$$

[a] Figure from Rosenzweig et al. 2022.

$$I(\mathbf{k},\omega) \propto \int \mathrm{d}t_1 \int \mathrm{d}t_2\, \mathrm{e}^{\mathrm{i}\,\omega(t_2-t_1)} \underbrace{G^{<}_{\mathbf{k}}(t_2,t_1)}_{\text{pumped (probe not considered)}} .$$

For Floquet-driven electron bands

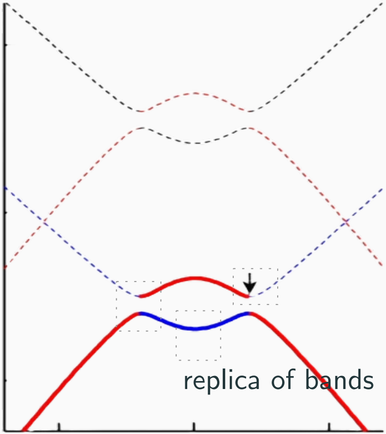
$$I(\mathbf{k},\omega) \sim \sum_{n,m} \left|\phi_n^{(m)}\right|^2 \delta(\omega - \varepsilon_{n\mathbf{k}}).$$

ARPES for Floquet-driven electron bands

Three effects of Floquet correction to ARPES spectra

In the figure:[a]

- Band replica: $H^{(0)} + m\hbar\omega$

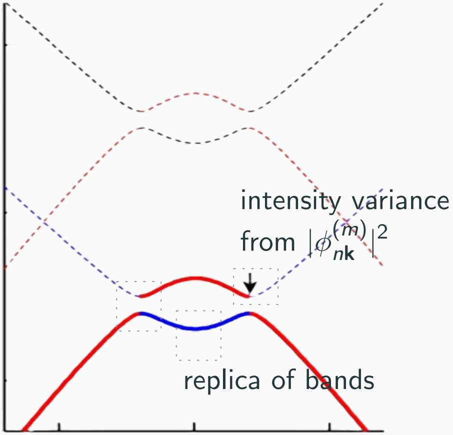


[a] Figure from Zhou et al. 2023

Three effects of Floquet correction to ARPES spectra

In the figure:[a]

- Band replica: $H^{(0)} + m\hbar\omega$
- Intensity peak: $\left|\phi_n^{(m)}\right|^2$



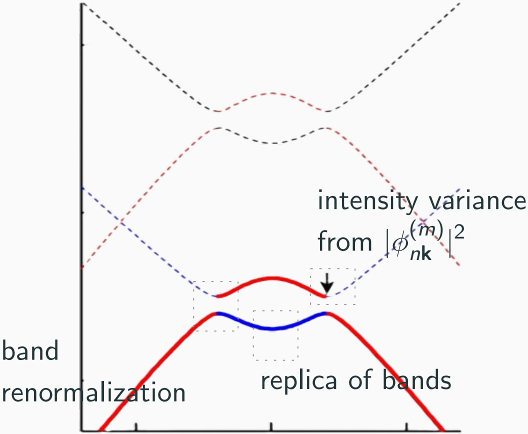
[a] Figure from Zhou et al. 2023

ARPES for Floquet-driven electron bands

Three effects of Floquet correction to ARPES spectra

In the figure:[a]

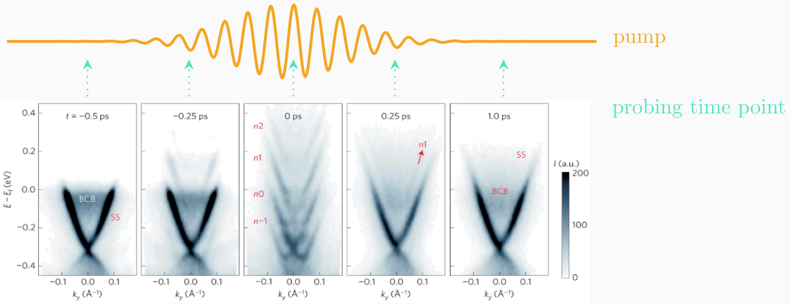
- Band replica: $H^{(0)} + m\hbar\omega$
- Intensity peak: $|\phi_n^{(m)}|^2$
- Band renormalization: $H^{(m\neq 0)}$



[a] Figure from Zhou et al. 2023

ARPES for Floquet-driven electron bands

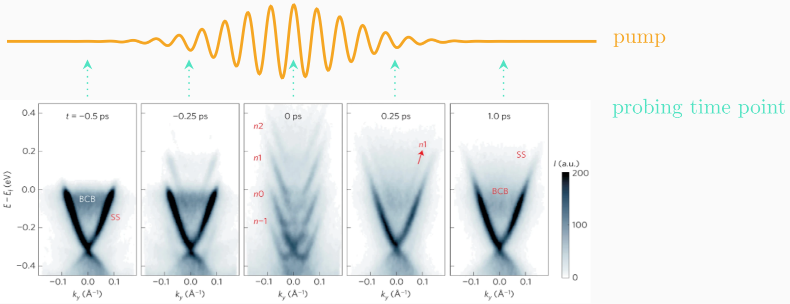
Probe at different stages of pump (Mahmood et al. 2016)



Probe before the start of pump nothing happens to bands

ARPES for Floquet-driven electron bands

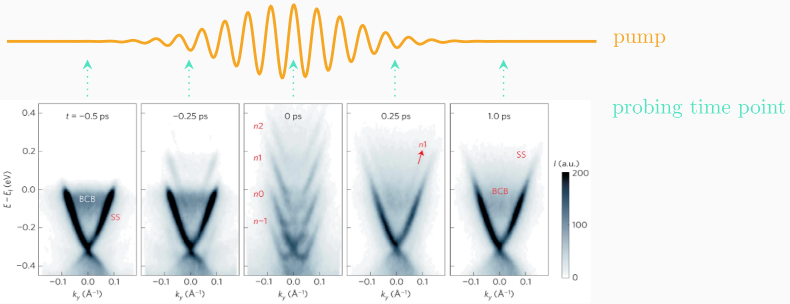
Probe at different stages of pump (Mahmood et al. 2016)



Probe at the start of pump mild Floquet features

ARPES for Floquet-driven electron bands

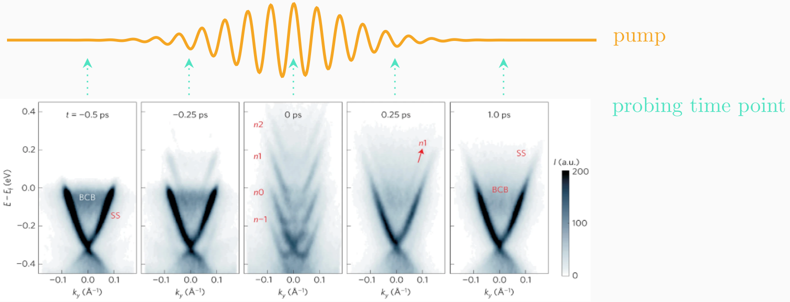
Probe at different stages of pump (Mahmood et al. 2016)



Probe at the middle of pump Floquet

ARPES for Floquet-driven electron bands

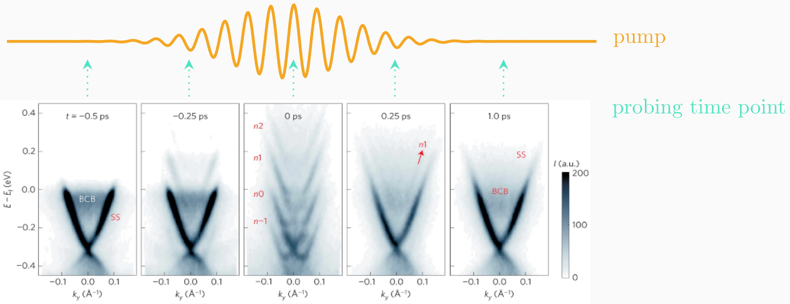
Probe at different stages of pump (Mahmood et al. 2016)



Probe at the tail of pump mild Floquet features, plus states excited to conduction bands

ARPES for Floquet-driven electron bands

Probe at different stages of pump (Mahmood et al. 2016)

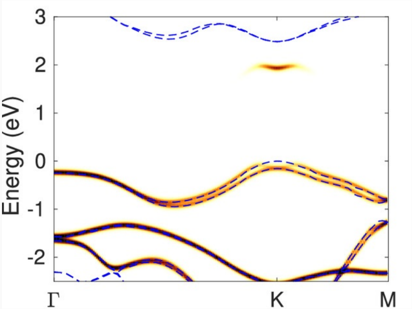


Probe after pump excited states

Self-driven Floquet effect

Even after pump ends we may still see Floquet effects...

Floquet by exciton excited by pump – self-driven Floquet effects (Chan et al. 2023)



Dotted line: band structure without pumping

Discussion

- Floquet quasienergies and quasi-stationary states
- They are “resummation” of external field’s perturbation
- They can be seen!

References

Chan, Y.-H. et al. (2023). “Giant self-driven exciton-Floquet signatures in time-resolved photoemission spectroscopy of MoS_2 from time-dependent *GW* approach”. In: *Proceedings of the National Academy of Sciences* 120.32, e2301957120.

Mahmood, Fahad et al. (2016). “Selective scattering between Floquet–Bloch and Volkov states in a topological insulator”. In: *Nature Physics* 12.4, pp. 306–310.

References ii

Rosenzweig, Philipp et al. (2022). "Surface charge-transfer doping a quantum-confined silver monolayer beneath epitaxial graphene". In: *Physical Review B* 105.23, p. 235428.

Zhou, Shaohua et al. (2023). "Pseudospin-selective Floquet band engineering in black phosphorus". In: *Nature* 614.7946, pp. 75–80.