Floquet physics

Periodic driving, formalism, and spetroscopy

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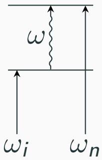
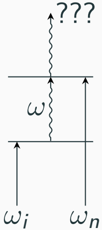
Introduction

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

- Floquet quasienergies and quasi-stationary states
- Relation with time-dependent perturbation theory and rotating wave approximation (RWA)?
- Floquet correction to ARPES

The Floquet formalism

Quasi-stationary states and quasienergies

Periodically driven Hamiltonian: quasi-eigensystem

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

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

- Time evolution of arbitrary states $\Leftrightarrow \{\,|\psi_n(t)\rangle\}$
- $\{\psi_n(t)\}$: **quasi-stationary states**
- $\{\varepsilon_n\}$: **quasienergies** (c.f. crystal momentum)

Our task How to find $\{\psi_n(t)\}$ and $\{\varepsilon_n\}$?

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

Floquet effective Hamiltonian

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

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

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

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

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

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

- Each "element" is a matrix on $\mathcal{H}$
- $H^{(0)}$: H without driving; $H^{(m)}$: driving with frequency $m\omega$
- H^{Floquet} is on the extended Hilbert space $\mathcal{H} \otimes \{m = \dots, -1, 0, 1, \dots\}$

Floquet Brillouin zone

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

$$\underbrace{e^{-\mathrm{i}(\varepsilon_n/\hbar+m\omega)t}\,\underbrace{e^{\mathrm{i}\,m\omega t}\,|\Phi_n(t)\rangle}_{\text{still periodic!}}}_{\text{both } \varepsilon_n \text{ and } \varepsilon_n + m\hbar\omega \text{ are its quasienergies}}$$

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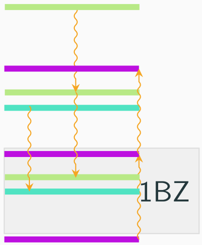


So only *one* energy Brillouin zone is needed.

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

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

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)

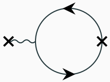
$$
\begin{tikzcd}
\text{Keldysh, etc.} \arrow[r, "T=0"] & \text{TD-PT} \arrow[r, "\text{periodic, no cutoff}"] \arrow[d] \arrow[rr, bend right] & \text{Floquet} \arrow[r, "{\text{small } \Omega, \Delta}"] & \text{RWA} \\
& \text{various cutoff schemes} & &
\end{tikzcd}
$$

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

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



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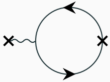
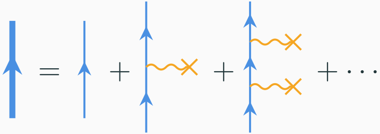


And when we sum over all of them...



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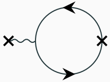
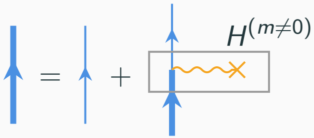


H^{Floquet} is the non-equilibrium self-energy



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Example: first-order PT = Lindhard response function

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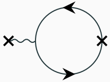

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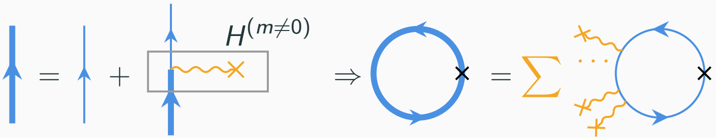


In the full H^{Floquet}, automatically all PT terms are considered!

Floquet formalism v.s. RWA

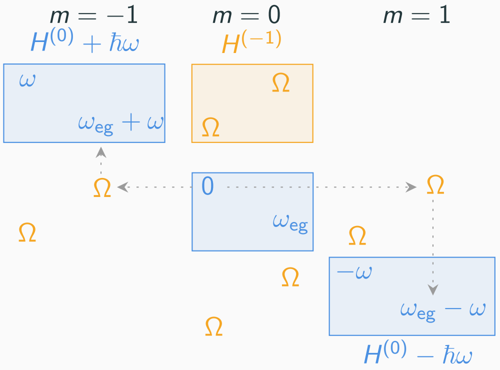
$$
\begin{array}{ccc}
m=-1 & m=0 & m=1 \\
H^{(0)}+\hbar\omega & H^{(-1)} & \\
\end{array}
$$

$$
\left(\begin{array}{cc|cc|cc}
\omega & & & \Omega & & \\
 & \omega_{\text{eg}}+\omega & \Omega & & & \\
\hline
 & \Omega & 0 & & & \Omega \\
\Omega & & & \omega_{\text{eg}} & \Omega & \\
\hline
 & & & \Omega & -\omega & \\
 & & \Omega & & & \omega_{\text{eg}}-\omega
\end{array}\right)
$$

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

From Floquet to RWA

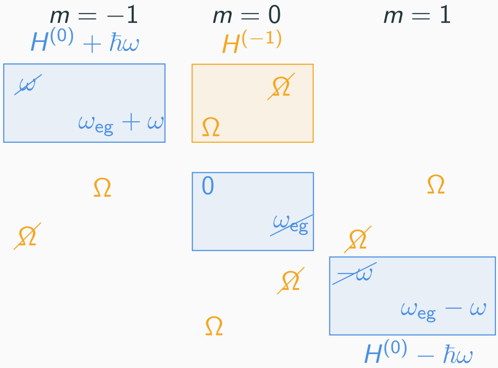
Floquet formalism v.s. RWA



From Floquet to RWA

- Weak driving: first-order transitions from $|g^{m=0}\rangle$ dominates

Floquet formalism v.s. RWA



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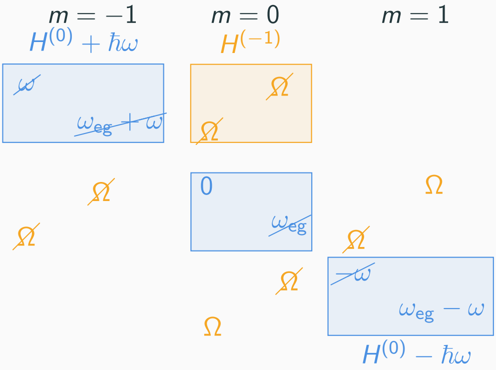
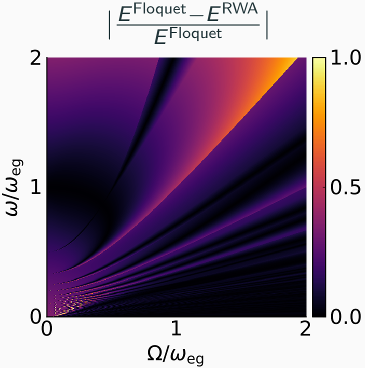


From Floquet to RWA

- Weak driving: first-order transitions from $|g^{m=0}\rangle$ dominates
- Near resonance: only $|e^{m=1}\rangle$ $(\omega_{eg} - \omega)$ matters

Floquet formalism v.s. RWA

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



From Floquet to RWA

- Weak driving: first-order transitions from $|g^{m=0}\rangle$ dominates
- Near resonance: only $|e^{m=1}\rangle$ $(\omega_{\mathrm{eg}} - \omega)$ matters

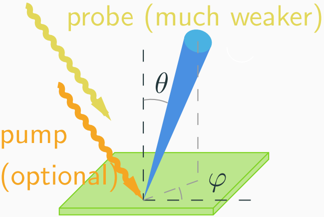
Indeed when $\Omega/\omega_{\mathrm{eg}} \lesssim 0.5$, $\omega/\omega_{\mathrm{eg}} \sim 1$, RWA works the best!

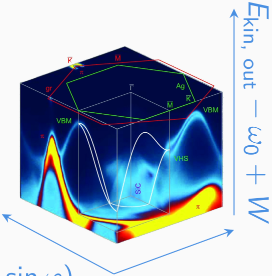
ARPES of Floquet systems

Are Floquet states “real”?

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

[a]Figure from Rosenzweig et al. 2022.





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

$$I_{\text{Chan et al. 2023}}(\mathbf{k}, \omega) \propto \underbrace{\int \mathrm{d}t_1 \int \mathrm{d}t_2\, \mathrm{e}^{\mathrm{i}\,\omega(t_2 - t_1)} \left| M^{\text{dipole}}_{\text{probe}} \right|^2 G^{\text{pumped},<}_{\mathbf{k}}(t_2, t_1)}_{\text{generalized Fermi golden rule for probing}}$$

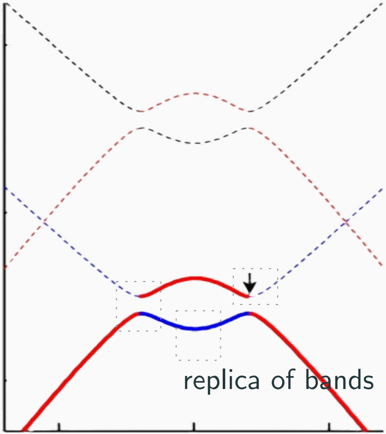
$$\Rightarrow \boxed{I_{\text{Floquet}}(\mathbf{k}, \omega) \sim \sum_{n,m} \left|\phi^{(m)}_{n\mathbf{k}}\right|^2 \delta(\omega - \varepsilon_{n\mathbf{k}}).}$$

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

ARPES for Floquet-driven electron bands

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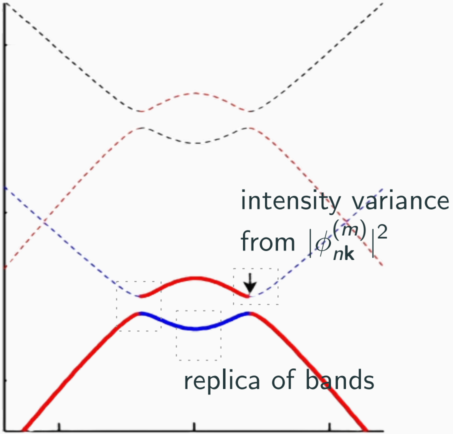
- Intensity peak: $\left|\phi_n^{(m)}\right|^2$



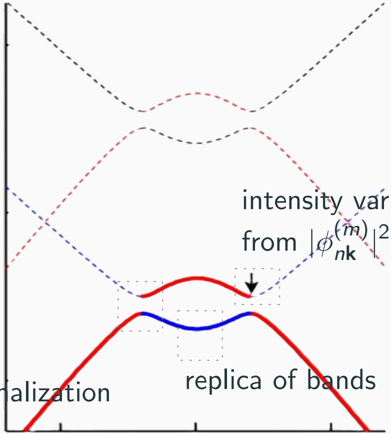
[a] Figure from Zhou et al. 2023

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: $H^{(m\neq 0)}$



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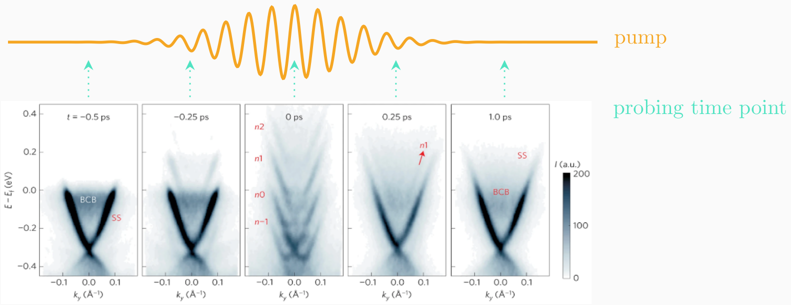
ARPES for Floquet-driven electron bands

Probe at different stages of pump in topological insulator Bi_2Se_3 (Mahmood et al. 2016)



Probe before the start of pump nothing happens to ARPES spectrum

ARPES for Floquet-driven electron bands

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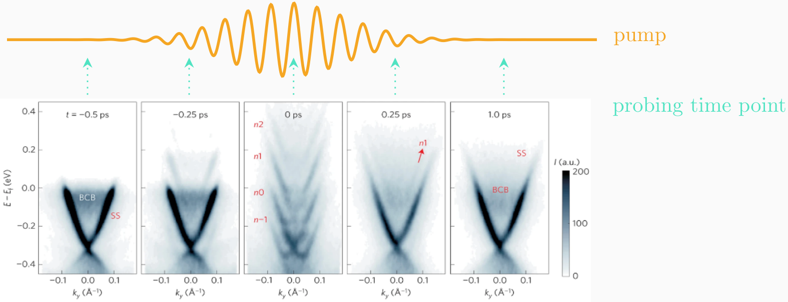

Probe at the start of pump mild Floquet features

ARPES for Floquet-driven electron bands

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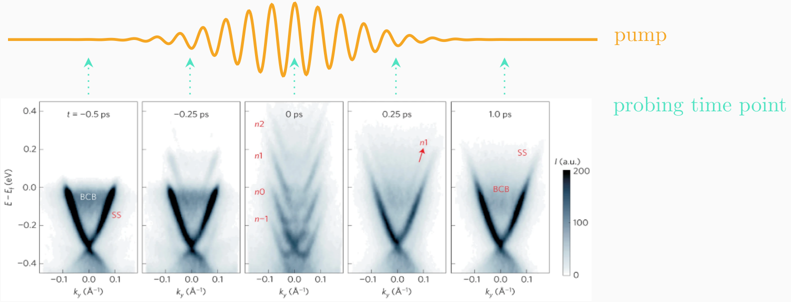

Probe at the middle of pump Floquet

ARPES for Floquet-driven electron bands

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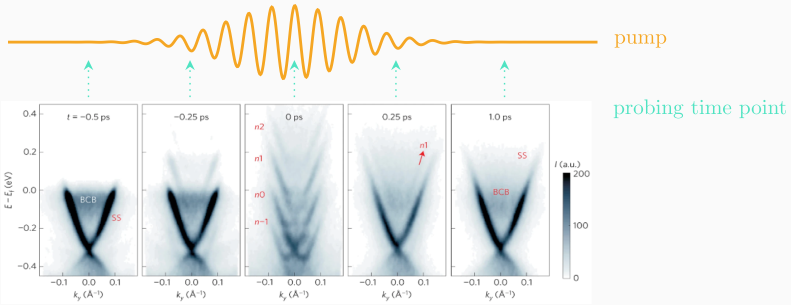
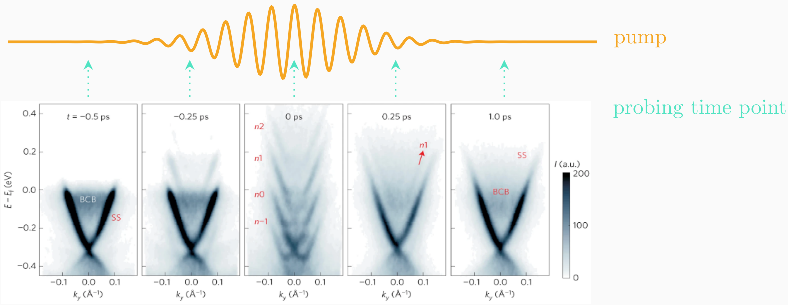


Probe at the tail of pump mild Floquet features, plus signatures from usually unoccupied states

ARPES for Floquet-driven electron bands

Probe at different stages of pump in topological insulator Bi_2Se_3 (Mahmood et al. 2016)

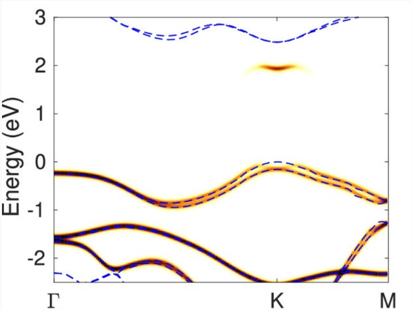


Probe after pump signatures from usually unoccupied states

Self-driven Floquet effect

Even after pump ends we may still see Floquet effects for excitonic materials like MoS_2...

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 by ARPES!

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.

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