Engineering quantum materials through light-matter interaction

Topics: 1 nonequilibrium dynamics from two-temperature models to Higgs spectroscopy
2 dressed states during a pulse: Floquet engineering
3 from Floquet to cavity engineering

Brief history: (incomplete!)

2103.14888
→ Colloquium to appear in Rev. Mod. Phys.

- short laser pulses thanks to "chirped pulse amplification" (Strickland & Mourou 1985, Nobel Prize 2018)
- ca. 1990: pump-probe experiments, e.g., on elemental metals/superconductors (Brorson et al., PRL 1990)
- photoinduced phase transitions (Koshihara et al., PRB 1990)
- ultrafast spin dynamics, e.g., in ferromagnetic Ni (Beaurepaire et al., PRB 1996)
- photoinduced structural phase transitions, e.g., in VO_2 (Cavalleri et al., PRL 2001)
- until mid 2000s mostly driving with optical frequencies.
  infrared laser (Ti:Sa) $\lambda = 800$ nm, $\hbar\omega = 1.5$ eV
  → typical energy scales for electronic dipole transitions
- new development: mid-infrared laser pulses
  → drive phonons resonantly
  e.g. - Rini et al., Nature 2007 lattice-controlled metal-insulator transition
  - Först et al., Nat. Phys. 2011
  - Subedi et al., PRB 2014 } "nonlinear phononics"
  → importance of phonon nonlinearities
  → light-induced superconducting-like optical properties across a range of materials (cuprates, fullerides, organic conductors)
  but no fully microscopic explanation yet.
- theory-driven conceptual development: 'Floquet engineering'
  = coherent dressing of electronic structure through periodic driving
  → Floquet topological insulators (Oka & Aoki, PRB 2009)

2003.08252
Rudner & Lindner,
"The Floquet Engineer's handbook"

1 Nonequilibrium dynamics from two-temperature models to Higgs spectroscopy

1.1 From frequency domain to time domain

Reminder: traditional spectroscopy works in frequency domain and relies on linear-response theory

$$H(t) = H_0 + H_1(t)$$

$$H_1(t) = -\int d^3r \, \underbrace{\delta h(\vec{r},t)}_{\text{e.g., magnetic field}} \, \underbrace{A(\vec{r})}_{\text{e.g., Pauli spin matrices}}$$

→ time-dependent perturbation theory gives

$$\langle B(\vec{r},t)\rangle = \langle B(\vec{r})\rangle_{eq} + \delta\langle B(\vec{r},t)\rangle$$

B = observable that we are interested in

and $\delta\langle B(\vec{q},\omega)\rangle = \chi^R_{BA}(\vec{q},\omega)\, \delta h(\vec{q},\omega)$ with

dynamical susceptibility $\chi^R_{BA}(\vec{q},\omega) \equiv \int dt \int d^3r \, e^{i\vec{q}\vec{r}} e^{i(\omega+i0^+)t} \times \chi^R_{BA}(\vec{r},t)$

with the retarded commutator

$$\chi^R_{BA}(\vec{r}-\vec{r}',t-t') \equiv \frac{i}{\hbar}\langle[B(\vec{r},t),A(\vec{r}',t')]\rangle_{eq}\,\Theta(t-t')$$

(1) response of observable B to field that couples to our system via A (A and B are both operators that belong to the system of interest)

② $\langle [B,A] \rangle_{eq}$ is an equilibrium (= intrinsic) property of the system

③ Linearity: response at momentum $\vec{q}$ and frequency ω is only to field at same $\vec{q}, \omega$; independent of response at other $\vec{q}', \omega'$

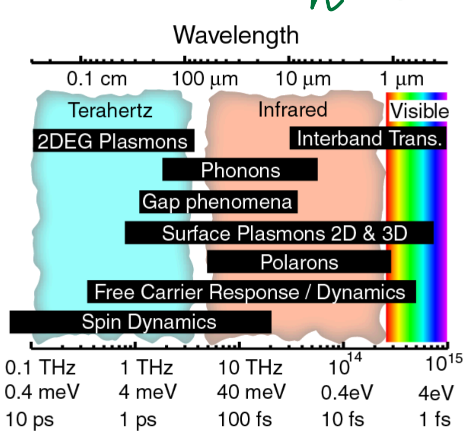
→ very useful techniques to investigate materials, <u>but</u>: frequency resolution often an issue in practice, especially at low frequencies ("emergent low-energy properties" like magnons)

can be hard to disentangle

→ Complementarity of frequency (energy) and time

$$\hbar = 0.658 \text{ eV} \cdot \text{fs}$$

(electron volts times femtoseconds)

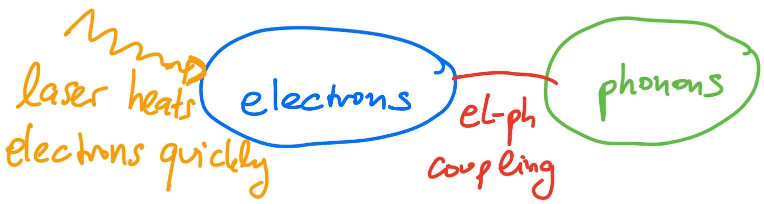


→ things that are hard to measure in the frequency domain can be easier to measure in the time domain!

1.2 Simplest dynamics in the time domain: two-temperature model

Consider electrons in a metal coupled to a phonon bath.

<u>Idea:</u> two subsystems that are each in a local thermal equilibrium, but <u>not</u> in a global equilibrium.



$\longrightarrow$ two-temperature model (Anisimov 1974, Allen 1987)

<u>Formal derivation:</u> semiclassical Boltzmann kinetic equations

$$H = \sum_k \epsilon_k \underbrace{c_k^+ c_k}_{\equiv \hat{n}_k} + \sum_Q \omega_Q \underbrace{b_Q^+ b_Q}_{\equiv \hat{N}_Q} \underbrace{- \sum_{kQ} M_Q \, c_{k+Q}^+ c_k (b_Q + b_{-Q}^+)}_{\text{el-ph coupling with strength } M_Q}$$

$c^{(+)}$ = fermionic } operators
$b^{(+)}$ = bosonic }

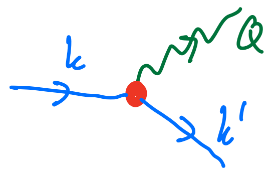
kinetic eqns.: (assume "well-defined quasiparticles")

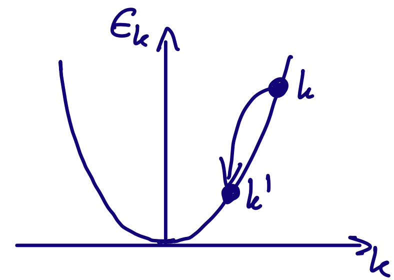
$$\frac{\partial n_k}{\partial t} = -2\pi \sum_{k',Q} \delta(k-k'-Q)\,|M_Q|^2 \Big\{ n_k(1-n_{k'}) \big[(N_Q+1)\,\delta(\epsilon_k-\epsilon_{k'}-\omega_Q) + N_Q\,\delta(\epsilon_k-\epsilon_{k'}+\omega_Q) \big] - (1-n_k)\,n_{k'} \big[(N_Q+1)\,\delta(\epsilon_k-\epsilon_{k'}+\omega_Q) + N_Q\,\delta(\epsilon_k-\epsilon_{k'}-\omega_Q) \big] \Big\}$$

$$\frac{\partial N_Q}{\partial t} = -4\pi \sum_{k,k'} \delta(k-k'-Q)\,|M_Q|^2 \Big\{ n_k(1-n_{k'}) \big[N_Q\,\delta(\epsilon_k-\epsilon_{k'}+\omega_Q) - (N_Q+1)\,\delta(\epsilon_k-\epsilon_{k'}-\omega_Q) \big] \Big\}.$$

Pictorially: $n_k(1-n_{k'})(N_Q+1)\,\delta(\epsilon_k-\epsilon_{k'}-\omega_Q)\,\delta(k-k'-Q)$

Diagram





scattering into lower-lying state by emission of a phonon

... and analogously the scattering-up process with phonon absorption.

Idea: simplify by making temperature ansatz:

$$n_k(t) = \frac{1}{1+e^{\epsilon_k/k_B T_e(t)}} \qquad T_e = \text{electronic temperature}$$

$$N_Q(t) = \frac{1}{e^{\omega_Q/k_B T_L(t)}-1} \qquad T_L = \text{lattice temperature}$$

Using this and computing the rates of energy exchange one can show that

$$\frac{\partial T_e}{\partial t} = \gamma_T\,(T_L - T_e)$$

$$\gamma_T \cong \frac{3\lambda\langle\omega^2\rangle}{\pi k_B T_e}$$

"Eliashberg function"

$$\alpha^2F(\Omega) \equiv \frac{2}{N(\epsilon_F)}\sum_{kk'}|M_Q|^2\delta(\omega_Q-\Omega) \times\delta(\epsilon_k-\epsilon_F) \times\delta(\epsilon_{k'}-\epsilon_F)$$

with $$\lambda\langle\omega^2\rangle \equiv 2\int_0^\infty d\Omega\,\frac{\alpha^2F(\Omega)}{\Omega}\,\Omega^2$$

→ relaxation rate of hot electrons after laser excitation provides information about the Eliashberg function, which for instance determines superconducting critical temperatures in conventional superconductors.
(measured using time-resolved photoemission spectroscopy)

1.3 Dynamics of ordered states: Higgs spectroscopy.

So far: change of distributions of electrons and phonons in a fixed band structure.

Now: What if the band structure itself becomes a dynamical quantity?

How can this happen? When there is a time-dependent self-energy. Simplest case: the self-energy is instantaneous.

$$\Sigma(t-t') \propto \delta(t-t')$$

$$\Leftrightarrow \Sigma(\omega) \propto \mathrm{const}(\omega)$$

"static" mean-field theory.

Example: BCS theory of superconductivity.

Attractive Hubbard model:

$$H = \sum_{k\sigma} \epsilon_{k\sigma} c^+_{k\sigma} c_{k\sigma} - U \sum_i c^+_{i\uparrow} c_{i\uparrow} c^+_{i\downarrow} c_{i\downarrow}$$

supports an s-wave superconducting solution with mean-field BCS Hamiltonian

$$H_{BCS} = \sum_{k\sigma} \underbrace{\epsilon_{k\sigma}}_{\epsilon_{k\uparrow} = \epsilon_{k\downarrow} \equiv \epsilon_k} c^+_{k\sigma} c_{k\sigma} - U \sum_{kk'} c^+_{k\uparrow} c^+_{-k\downarrow} \langle c_{-k'\downarrow} c_{k'\uparrow} \rangle + h.c.$$

$$= \sum_k \underline{\psi}^+_k \underline{\underline{h}}_{BCS}(k) \underline{\psi}_k$$

Nambu spinor $\underline{\psi}^+_k \equiv (c^+_{k\uparrow}, c_{-k\downarrow})$

$\underline{\psi}_k \equiv (\underline{\psi}^+_k)^+$

Matrix Hamiltonian $\underline{\underline{h}}_{BCS}(k) = \begin{bmatrix} \epsilon_k & \Delta \\ \Delta^* & -\epsilon_{-k} \end{bmatrix}$

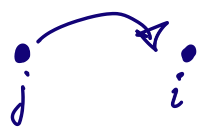
Gap function ("self-energy") $\Delta \equiv U \sum_{k'} \underbrace{\langle c_{-k'\downarrow} c_{k'\uparrow} \rangle}_{\equiv f_{k'}}$. "anomalous expectation value"

(i) Equilibrium: The gap Δ is determined self-consistently by solving the self-consistency equation

$$\Delta = U \sum_{k'} \frac{\Delta}{\sqrt{\epsilon_{k'}^2 + \Delta^2}} \tanh\left(\frac{\beta\sqrt{\epsilon_{k'}^2+\Delta^2}}{2}\right)$$

with inverse temperature $\beta = 1/k_B T$.

(ii) <u>Nonequilibrium:</u> Starting from the equilibrium solution we propagate forward in time under an external driving field. Light-matter coupling is modelled through "minimal coupling" $\vec{p} \longrightarrow \vec{p} - e\vec{A}$, which in the context of lattice electrons is achieved through "Peierls substitution":



$t_{ij}\, c_i^+ c_j$ hopping from j to i

is dressed as

$$t_{ij}\, e^{\frac{ie}{\hbar}\int_{r_j}^{r_i} d\vec{\ell}\cdot\vec{A}(\vec{\ell},t)}$$

Here $\vec{A}(\vec{r},t)$ is the vector potential (gauge field).

Optical laser: focus on electric field $\vec{E} = -\frac{\partial \vec{A}}{\partial t}$, wavelength $\lambda = 800$ nm $\Rightarrow$ atomic length scales

$\Rightarrow$ can neglect spatial dependence "dipole approximation" $e^{i\vec{q}\cdot\vec{r}} \approx 1$ for $\vec{q} = \frac{2\pi}{\lambda} \approx 0$

$\Rightarrow$ Peierls substitution amounts to the replacement $\vec{k} \rightarrow \vec{k} - e\vec{A}$.

$$H_{BCS}(t) = \sum_k \underline{\psi}_k^+ \begin{bmatrix} \epsilon_{k-eA(t)} & \Delta(t) \\ \Delta^*(t) & -\epsilon_{-k-eA(t)} \end{bmatrix} \underline{\psi}_k .$$

<u>Note:</u> t-dependence in $A(t)$ is from external laser field. t-dependence in $\Delta(t)$ is from internal dynamics reacting to the external field.

Now: Need equation of motion (EOM) for dynamics. We will use the Heisenberg picture (time-dependent operators):

$$i\partial_t \underline{\underline{G}}^<(k,t,t) = [\underline{\underline{h}}_{BCS}(k,t), \underline{\underline{G}}^<(k,t,t)]$$

with $\underline{\underline{G}}^<(k,t,t) := i\langle \underline{\Psi}_k^+(t)\, \underline{\Psi}_k(t)\rangle$.

Idea (Anderson, Phys. Rev. 112, 1900 (1958)):

introduce Anderson pseudospin:

$$\vec{\sigma}_k \equiv \frac{1}{2}\underline{\Psi}_k^+ \cdot \vec{\underline{\underline{\tau}}} \cdot \underline{\Psi}_k \qquad \text{with Pauli matrices } \vec{\underline{\underline{\tau}}} = \begin{pmatrix} \underline{\underline{\tau}}^x \\ \underline{\underline{\tau}}^y \\ \underline{\underline{\tau}}^z \end{pmatrix}$$

one can show that this leads to EOMs

$$(*) \quad \boxed{\partial_t\langle\vec{\sigma}_k\rangle = 2\,\vec{b}_k \times \langle\vec{\sigma}_k\rangle} \quad \text{with } \vec{b}_k = \begin{pmatrix} \Delta' \\ \Delta'' \\ \epsilon_{k-eA}+\epsilon_{k+eA} \end{pmatrix}.$$

"pseudospin precession under pseudomagnetic field"

For example:

$$\sigma_k^x = \frac{1}{2}\left(c_{k\uparrow}^+, c_{-k\downarrow}\right)\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}\begin{pmatrix} c_{k\uparrow} \\ c_{-k\downarrow}^+ \end{pmatrix}$$

$$= \frac{1}{2}\left(c_{k\uparrow}^+ c_{-k\downarrow}^+ + c_{-k\downarrow} c_{k\uparrow}\right)$$

$$\sigma_k^y = \frac{1}{2}\left(c_{k\uparrow}^+, c_{-k\downarrow}\right)\begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}\begin{pmatrix} c_{k\uparrow} \\ c_{-k\downarrow}^+ \end{pmatrix}$$

$$= \frac{1}{2}\left(-i\, c_{k\uparrow}^+ c_{-k\downarrow}^+ + i\, c_{k\downarrow} c_{k\uparrow}\right)$$

$$\Rightarrow \langle\sigma_k^x\rangle = \frac{1}{2}\left(f_k^* + f_k\right) = \mathrm{Re}\, f_k = f_k'$$

$$\langle\sigma_k^y\rangle = \frac{1}{2}\left(-i f_k^* + i f_k\right) = \mathrm{Im}\, f_k = f_k''$$

$$\sigma_k^z = \frac{1}{2}\left(c_{k\uparrow}^+ c_{k\uparrow} - \underbrace{c_{k\downarrow} c_{k\downarrow}^+}_{-c_{k\downarrow}^+ c_{k\downarrow}+1}\right)$$

$$\left\{\begin{array}{l} \{c, c^+\} = 1 \\ cc^+ + c^+c = 1 \\ cc^+ = 1 - c^+c \end{array}\right.$$

$$= \frac{1}{2}\left(\underbrace{c_{k\uparrow}^+ c_{k\uparrow} + c_{-k\downarrow}^+ c_{-k\downarrow}}_{=2\bar{n}_k} - 1\right)$$

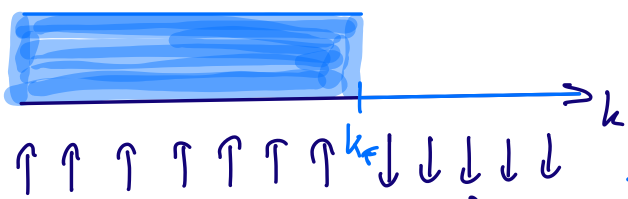
So that (*) implies

$$\partial_t f_k' = 2\,\Delta'' \cdot \tfrac{1}{2}(2n_k-1) - 2\,\frac{\epsilon_{k-eA}+\epsilon_{k+eA}}{2}\, f_k''$$

$$\partial_t f_k'' = 2\,\frac{\epsilon_{k-eA}+\epsilon_{k+eA}}{2}\, f_k' - 2\,\Delta' \cdot \tfrac{1}{2}(2n_k-1)$$

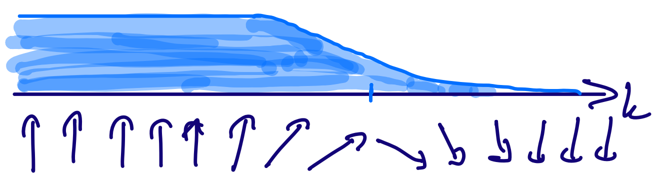
$$\underbrace{\partial_t \tfrac{1}{2}(2n_k-1)}_{=\partial_t n_k} = 2\Delta' f_k'' - 2\,\Delta'' f_k' .$$

Intuitive picture of Anderson pseudospin: spins in k space



normal state (metal)

$\Rightarrow$ only σ^z component, $\uparrow \hat{=}$ filled, $\downarrow =$ empty



BCS state $\Rightarrow$ in-plane component near k_F signals SC condensate

Now back to dynamics: Pseudospin precession is governed by $\vec{b}_k = \begin{pmatrix} \Delta' \\ \Delta'' \\ \frac{\epsilon_{k-eA(t)} + \epsilon_{k+eA(t)}}{2} \end{pmatrix}$

$\Rightarrow$ particle-hole-symmetric by construction

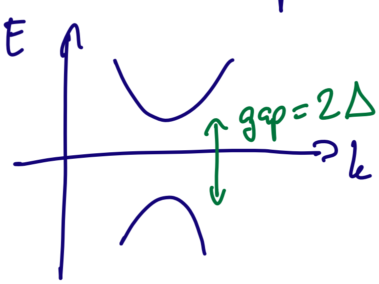
$\Rightarrow$ linear response vanishes since:

$$\frac{1}{2}\left(\epsilon_{k-eA} + \epsilon_{k+eA}\right) = \epsilon_k + \frac{1}{2}\frac{\partial \epsilon_k}{\partial k}\cdot\cancel{(-eA+eA)} + \frac{1}{2}\cdot\frac{1}{2}\frac{\partial^2 \epsilon_k}{\partial k^2}\left((eA)^2+(eA)^2\right)$$

$$= \epsilon_k + \frac{1}{2}\frac{\partial^2\epsilon_k}{\partial k^2} e^2 A^2$$

⟹ Superconductor couples to gauge field only at second order! (consequence of gauge invariance)

⟹ "Anderson pseudospin resonance" (Tsuji & Aoki, Phys. Rev. B 92, 064508 (2015))



$$(A(t))^2 = \Big(A_0 \underbrace{s(t)}_{\text{pulse shape}} \cos(\omega t)\Big)^2$$

$$= A_0^2\, s(t)^2 \underbrace{\cos^2(\omega t)}_{\text{oscillates at } 2\omega}$$

⟹ resonance condition

$$2\omega = 2\Delta$$

$$\omega = \Delta = \text{half the gap.}$$

→ Collective mode that is excited by this nonlinear process corresponds to "Higgs mode" (oscillation at 2Δ frequency) = Amplitude mode of the SC condensate.

→ "Higgs spectroscopy". Review by Shimano & Tsuji, Annu. Rev. 11, 103 (2020) of Cond. Mat. Phys.

So far:
- "hot electron dynamics"
- nonlinear response of collective mode in time domain spectroscopy

→ information about microscopic couplings and collective modes of the material itself.

But: Can we also change (control) materials properties with light?

2 Dressed states during a pulse: Floquet engineering

Q: What happens when electrons couple to a time-dependent laser field?

Simplest case: field is periodic in time, $f(t+T) = f(t)$.

Real-space analogy: spatially periodic potential ⇒ Bloch theorem
⇒ we can use quasi-momentum $\vec{k}$ to label eigenstates
(good quantum number)
⇒ similar construction in time-energy space?

Time-dependent Schrödinger equation:

$$i\hbar \frac{d}{dt} |\psi(t)\rangle = H(t) |\psi(t)\rangle \quad , \quad H(t+T) = H(t).$$

Floquet theorem: we can define Floquet states and label them with quasi-energy ε (good quantum number)

Floquet states are stationary states of the stroboscopic ("Floquet") time evolution operator

$$U(T) \equiv \mathcal{T} e^{-\frac{i}{\hbar}\int_0^T dt' H(t')}$$

$\mathcal{T}$: time ordering

2.1 Frequency-(extended) space formulation

Reminder: the Bloch theorem allows us to convert the problem of solving a differential equation

$$\left(-\frac{\hbar^2\nabla^2}{2m} + V(x)\right)\psi(x) = E\,\psi(x)$$

into an eigenvalue problem $\left(V(x) = \sum_G e^{iGx} U_G\right)$

$$(***)\quad \frac{\hbar^2}{2m}(k+G)^2\, c_G(k) + \sum_{G'} U_{G-G'}\, c_{G'}(k) = \varepsilon_k\, c_G(k)$$

with Bloch wave functions (same periodicity as lattice!)

$$(**)\quad u_k(x) = \sum_G c_G(k)\, e^{iGx}$$

$u_k(x \pm a) = u_k(x)$
a = lattice constant

and "full" wave function

$$(*)\quad \psi_k(x) = e^{ikx}\, u_k(x) = \text{plane wave} \times \text{Bloch function}$$

with quasi-momentum k and reciprocal lattice vector G.

Real-time construction:

$$(*)\quad |\psi_n(t)\rangle = e^{-i\varepsilon_n t/\hbar}\, |\phi_n(t)\rangle, \quad |\phi_n(t+T)\rangle = |\phi_n(t)\rangle$$

(**) $|\phi_n(t)\rangle = \sum_m e^{-im\omega t} |\phi_n^{(m)}\rangle$

$\Rightarrow$ ϕ_n can be decomposed into harmonics of the drive frequency $\omega = \frac{2\pi}{T}$.

$|\phi_n^{(m)}\rangle \equiv$ m-th Fourier coefficient of $|\phi_n(t)\rangle$

Remark: the wavefunction $|\psi_n(t)\rangle$ is normalized at every t; but the Fourier coefficient are in general <u>not</u> normalized, i.e., typically $\langle \phi_n^{(m)} | \phi_n^{(m)} \rangle < 1$. (same is true for $C_G(k)$ in the Bloch-wave expansion.)

Goal: Find algebraic equation analogous to (***).

Using the Floquet ansatz (*) the Schrödinger equation becomes $\left[\varepsilon_n + i\hbar \frac{d}{dt}\right] |\phi_n(t)\rangle = H(t) |\phi_n(t)\rangle$.

With (**) and the Fourier decomposition of $H(t)$,

$$H(t) = \sum_m e^{-im\omega t} H^{(m)}$$

we obtain $\left[\varepsilon_n + m\hbar\omega\right] |\phi_n^{(m)}\rangle = \sum_{m'} H^{(m-m')} |\phi_n^{(m')}\rangle$.

We can cast this into the form of an eigenvalue problem by creating a vector

$$\underline{\varphi}_n = \left\{ |\phi_n^{(m)}\rangle, \ m = -\infty, \dots, \infty \right\}$$

and writing

$$\underline{\underline{H}}\,\underline{\varphi}_n = \varepsilon_n \underline{\varphi}_n, \qquad \underline{\underline{H}} \equiv \begin{bmatrix} \ddots & H^{(-1)} & H^{(-2)} & \cdot^{\cdot^{\cdot}} \\ H^{(1)} & H_0 - m\hbar\omega & H^{(-1)} & H^{(-2)} \\ H^{(2)} & H^{(1)} & H_0-(m-1)\hbar\omega & \\ \cdot^{\cdot^{\cdot}} & H^{(2)} & H^{(1)} & \ddots \end{bmatrix},$$

$$\underline{\varphi}_n = \begin{bmatrix} \vdots \\ |\phi_n^{(m)}\rangle \\ |\phi_n^{(m-1)}\rangle \\ \vdots \end{bmatrix}.$$

$$H_0 \equiv H^{(0)} = \frac{1}{T}\int_0^T dt\ H(t)$$ "dc" = time-averaged part of Hamiltonian.

Each entry in $\underline{\underline{H}}$ is itself a $d\times d$ matrix, with d = dimension of original Hilbert space.

$\Rightarrow$ we have simplified the solution of a time-periodic problem to the solution of an (infinite-dimensional) eigenvalue problem.

The time-periodic part of the Floquet state wavefunction is then obtained from

$$|\phi_n(t)\rangle = \underbrace{\mathcal{P}(\omega t)}_{\substack{\text{rectangular matrix} \\ \text{with blocks } e^{-im\omega t}\mathbb{1}_{d\times d}}} \underline{\varphi}_n \equiv \sum_m e^{-im\omega t} |\phi_n^{(m)}\rangle$$

<u>Overcompleteness:</u> Like in band structure theory, the Fourier space solution has redundancy. We can shift

each solution via $\varepsilon_n \to \varepsilon_n + m'\hbar\omega$ by integer multiples of $\hbar\omega$ without changing the physical state. $\Rightarrow$ "Floquet Brillouin zone" of width $\hbar\omega$

(reflects conservation of energy only modulo integer multiples of the photon energy)

2.2 Truncation of frequency space

What have we gained by the Floquet picture?

Consider $H(t) = H_0 + V(t)$, $V(t) = Ve^{i\omega t} + V^\dagger e^{-i\omega t}$

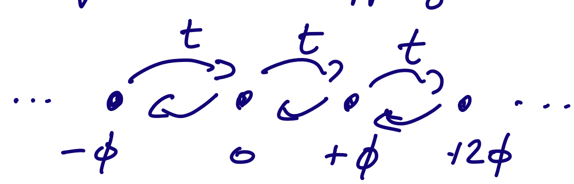
$\to$ purely harmonic drive that couples linearly to the system

$$\to H^{(-1)} = V, \quad H^{(1)} = V^\dagger, \quad H^{(\Delta m)} = 0 \text{ for } |\Delta m| > 1$$

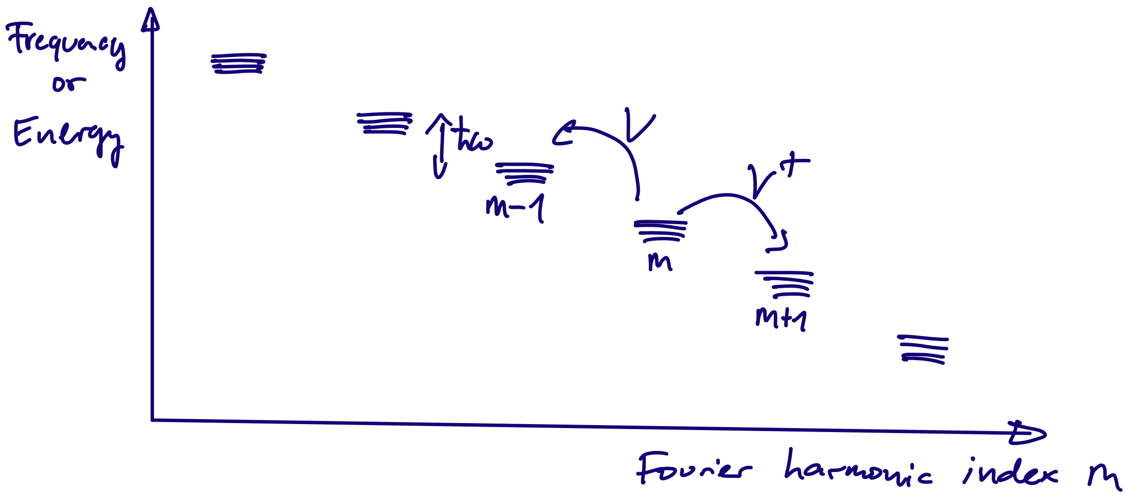
$\Rightarrow$ block-tridiagonal form

$$\underline{\underline{H}} = \begin{bmatrix} \ddots & V & 0 \\ V^\dagger & H_0 - m\hbar\omega & V \\ 0 & V^\dagger & \ddots \end{bmatrix}$$

$\Rightarrow$ closely analogous to hopping on a tight-binding lattice



with an energy offset $\Delta E = \phi$ between neighboring sites. (Physics of Bloch oscillations and dc Wannier-Stark effect.)



$\Rightarrow$ the temporally periodic potential with strength V hybridizes the "local" Hilbert spaces which are offset by multiples of $\hbar\omega$.

$\Rightarrow$ if we are interested in the physics within a certain energy window, we can often truncate the harmonics because higher-lying (lower) states will only contribute perturbatively as $\sim \frac{V^n}{n\hbar\omega}$ if their distance is $n\hbar\omega$.

$\Rightarrow$ in practice, introduce a cutoff and check convergence as the cutoff is varied.

2.3 Two elementary examples of Floquet engineering

How can periodic driving be used to change ("control with light") a band structure?

(a) Dynamical localization

Particle hopping in a 1D chain exposed to electromagnetic field:

$$\epsilon(k) = -2t_0 \cos(ka)$$

$$H = -\sum_n t_0 \, c_n^+ c_{n+1} + h.c. = \sum_k \epsilon(k) \, c_k^+ c_k$$

Peierls substitution: $t_0 \to t_n(t) = t_0 \, e^{i e a A(t)/\hbar}$

$$A(t) = \frac{E_0}{\omega} \cos(\omega t)$$

$\Rightarrow$ consider high-frequency limit $\omega \gg t_0$

$\Rightarrow$ effectively time-averaged Hamiltonian

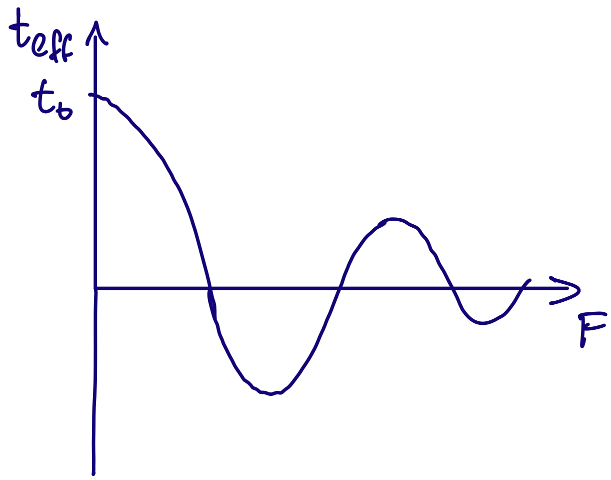
$$H_0 = \frac{1}{T} \int_0^T (-2t_0) \cos\left(\left(k - \frac{eE_0}{\hbar\omega} \cos(\omega t)\right) a\right) dt$$

$\Rightarrow$ $\int \cos(\cos)$ yields a Bessel function

$$\Rightarrow \quad H_0 = -\sum_n t_{eff} \, c_n^+ c_{n+1} + h.c.$$

with $\quad t_{eff} = t_0 \, J_0(F)$

$\quad F \equiv a e E_0 / (\hbar\omega)$ "Floquet parameter"



$|t_{eff}| \leq t_0$ due to $|J_0(x)| \leq 1$

$\Rightarrow$ reduction of effective hopping amplitude; in strong field it can even flip sign

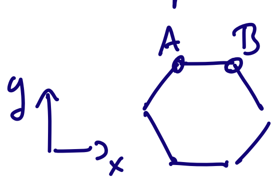
$\rightarrow$ "dynamical localization"

(Dunlap & Kenkre 1986; Bucksbaum et al. 1990)

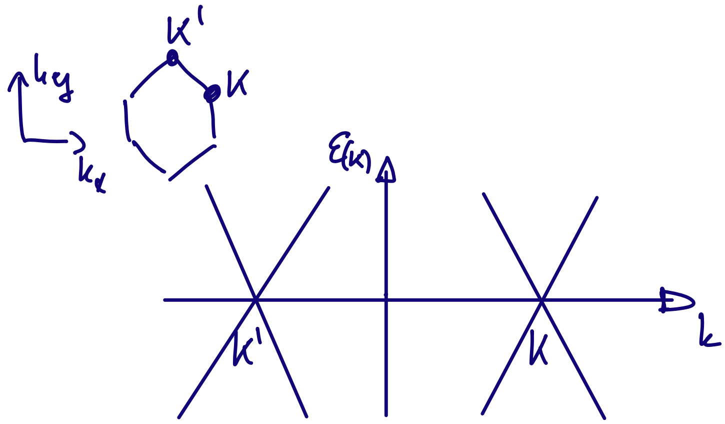
bond softening in hydrogen molecules under laser irradiation

(b) Gap opening in Dirac fermions under circularly polarized light: "Floquet topological insulator"

This idea was put forward by Oka & Aoki in 2009 and inspired by the "Haldane model" (1988): "modified honeycomb lattice"



regular hopping on honeycomb: Dirac cones at $K, K' = -K$ in hexagonal Brillouin zone



massless Dirac fermions: $\quad k = K^{(\prime)} + q$

$$H(k) \approx v_F \begin{bmatrix} 0 & k_x \pm i k_y \\ k_x \mp i k_y & 0 \end{bmatrix}$$

where $\pm$ refers to K (K')

How can one create a gap ($\hat{=}$ mass term)?

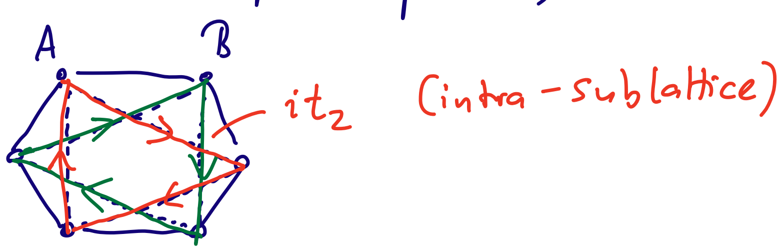
$\Rightarrow$ break one (or both) of two symmetries:

1) inversion

2) time reversal

case 1): introduce onsite potential $\varepsilon_A \neq \varepsilon_B$

case 2): introduce "effective magnetic field" via second neighbor hopping with complex phase; "chiral hopping"



$$\Rightarrow \quad H(\vec{k}) = H_0(\vec{k}) + \underbrace{M}_{=\varepsilon_A - \varepsilon_B} \sigma_z + 2t_2 \sum_i \sigma_z \sin(\vec{k}\cdot\vec{b}_i)$$

$\Rightarrow$ $t_2 = \pm \frac{M}{3\sqrt{3}}$ can close trivial M gap at one of the Dirac points

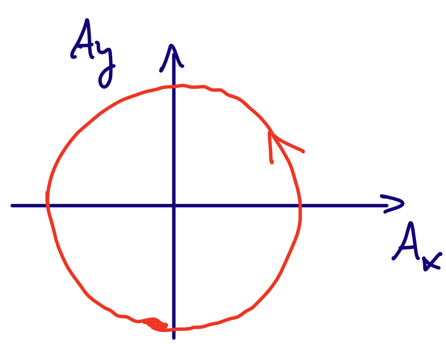
$\Rightarrow$ transition from trivial to Chern insulator

Q: Can one Floquet-engineer the t_2 term?

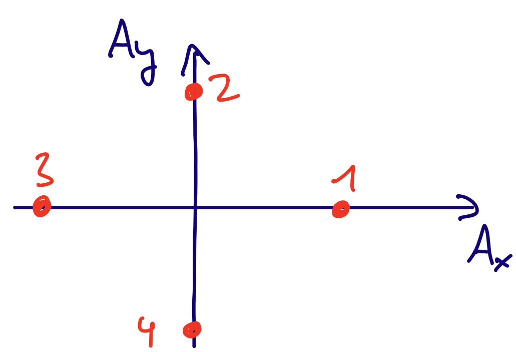
A: Yes! Need to break trs $\Rightarrow$ circularly polarized light.

<u>Two models:</u>

1. toy model: 4-step process where we rotate a vector potential over quarters of a driving period $\frac{T}{4}$.



$\Rightarrow$



Bloch Hamiltonian $\underline{\underline{H}}_n(\vec{k}) = v_F \left[\hbar \vec{k} - e A_0 \vec{e}_n\right] \cdot \vec{\sigma}$

in step $n \in \{1, 2, 3, 4\}$.

$\Rightarrow$ Floquet time evolution operator

$$U(\vec{k}, T) = U_4(\vec{k})\, U_3(\vec{k})\, U_2(\vec{k})\, U_1(\vec{k})$$

$$U_n(\vec{k}) = e^{-i H_n(\vec{k}) T/(4\hbar)}$$

At the Dirac point ($\vec{k} = 0$ here):

$$U(0,T) = e^{-i\phi\sigma_y}\, e^{-i\phi\sigma_x}\, e^{i\phi\sigma_y}\, e^{i\phi\sigma_x}$$

with $\phi \equiv e v_F A_0 T/(4\hbar)$.

High-frequency (small T) limit: expand exp's.

$$U(0,T) = 1 + \phi^2 [\sigma_x, \sigma_y] + \mathcal{O}(\phi^3)$$

$$= 1 + 2i\phi^2 \sigma_z \simeq e^{2i\phi^2\sigma_z}$$

Define $U(0,T) =: e^{-i H_{eff}(\vec{k}=0) T/\hbar}$

and $T = \frac{2\pi}{\omega}$

$$\Rightarrow \boxed{H_{eff}(\vec{k}=0) = \tilde{\Delta}\sigma_z, \quad \tilde{\Delta} \equiv -\frac{\pi (e v_F A_0)^2}{4\hbar\omega}}$$

- circularly polarized driving opens gap at Dirac point, gap $\propto (\text{field})^2 = \text{intensity}$ of laser.
- reversal of handedness of light $\Leftrightarrow$ reversal of gap. (sign of mass term)
- reversal of chirality of Dirac fermion also reverses the sign of mass term !!

  $\Rightarrow$ corresponds to it_2 hopping in Haldane model

(2) Dirac model with continuous drive

$$\vec{A}(t) = A_0 \begin{pmatrix} \cos \omega t \\ \sin \omega t \end{pmatrix}$$

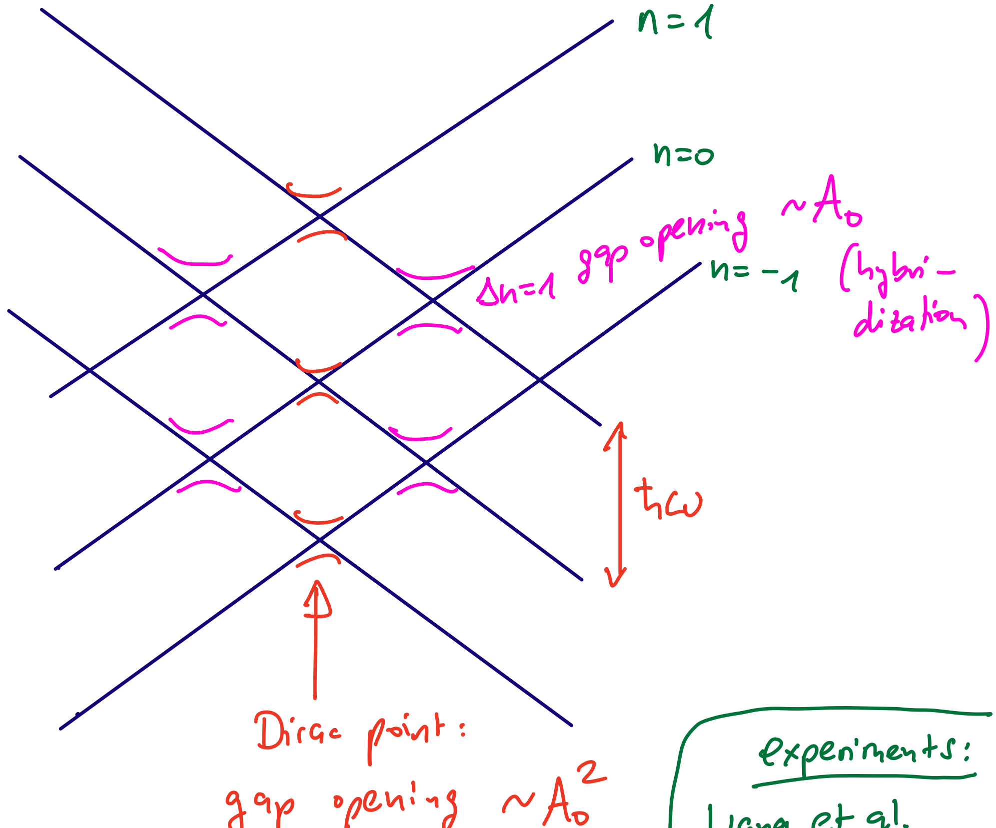
$$H(\vec{k},t) = v_F \left[\hbar \vec{k} - e\vec{A}(t)\right] \cdot \vec{\sigma}$$

$$= v_F \begin{bmatrix} 0 & \hbar k + eA_0 e^{i\omega t} \\ \hbar k^* + eA_0 e^{-i\omega t} & 0 \end{bmatrix}$$

$$k := k_x + i k_y$$

Floquet Hamiltonian: $$H_{mn} = \frac{1}{T}\int_0^T dt\, H(t) e^{i(m-n)\omega t} + m\,\delta_{mn} \hbar\omega \mathbb{1}$$

$$\Rightarrow \quad H_F = \begin{bmatrix} \ddots & & & & & & & \\ & \hbar\omega & \hbar v_F k & 0 & e v_F A_0 & & & \\ & \hbar v_F k^* & \hbar\omega & 0 & 0 & & & \\ & 0 & 0 & 0 & \hbar v_F k & 0 & e v_F A_0 & \\ & e v_F A_0 & 0 & \hbar v_F k^* & 0 & 0 & 0 & \\ & & & 0 & 0 & -\hbar\omega & \hbar v_F k & \\ & & & e v_F A_0 & 0 & \hbar v_F k^* & -\hbar\omega & \\ & & & & & & & \ddots \end{bmatrix}$$



experiments:
Wang et al. Science 2013
Mahmood et al. Nat. Phys. 2016
McIver et al. Nat. Phys. 2020

2nd order perturbation theory:

$$H_{eff} = H_0 + \frac{[H_{-1}, H_{+1}]}{\hbar\omega} + \mathcal{O}(A_0^4)$$

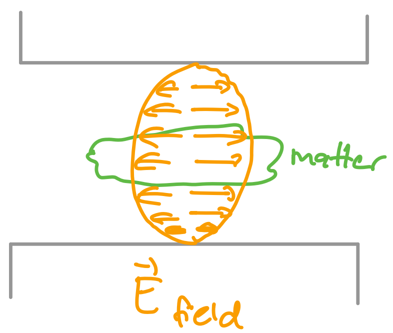
$$= H_0 + \boxed{\frac{e^2 v_F^2 A_0^2}{\hbar\omega}\sigma_z}$$

dynamically generated mass term, very similar to toy model.

3. From classical to quantum Floquet engineering in QED cavities

We can envision that "photon dressing effects" can also change materials properties even when classical EM fields are absent.

Cavity



Floquet

$$H = H_{matter} + H_{light-matter}[\vec{A}]$$

$$H = H_{matter} + \sum_{\vec{q}s} \omega_{\vec{q}}\, a^{\dagger}_{\vec{q}s} a_{\vec{q}s}$$

$$+ H_{light-matter}\Big[\vec{A} \to \hat{\vec{A}}(\vec{r}) = i \sum_{\vec{q}s} \sqrt{\frac{\hbar}{\epsilon\epsilon_0 V \omega_{\vec{q}}}}\, e^{i\vec{q}\cdot\vec{r}} \times$$

$$\times\, \hat{e}_{\vec{q}s}\left(a_{\vec{q}s} + a^{\dagger}_{-\vec{q}s}\right)\Big]$$

Floquet: Laser prepares photons in many-photon coherent state ("classical"); $\vec{A}$ has finite amplitude.

Cavity: $\hat{A} \propto (a + a^{+})$ can have zero amplitude (no macroscopic field) but still impact matter through its fluctuations.

Examples: Casimir effect, Purcell effect (enhancement of spontaneous emission)

Now: one example, namely quantum analogue of Floquet Chern insulator

X. Wang et al., PRB 99, 235156 (2019)

Single Dirac fermion:

$$H = \sum_k \begin{pmatrix} c^{+}_{A,k} \\ c^{+}_{B,k} \end{pmatrix} \overbrace{\begin{bmatrix} 0 & \gamma(\vec{k}-\vec{A}) \\ \gamma(\vec{k}-\vec{A})^{+} & 0 \end{bmatrix}}^{H(k)} \begin{pmatrix} c_{A,k} \\ c_{B,k} \end{pmatrix}$$

$$+ \sum_\lambda \omega_\lambda a^{+}_\lambda a_\lambda$$

$$\vec{A} = A_0 \sum_\lambda (\vec{e}_\lambda a_\lambda + \vec{e}^{\,*}_\lambda a^{+}_\lambda)$$

$$A_0 = \sqrt{\frac{\hbar}{\epsilon \epsilon_0 V \omega}}$$

$$\gamma(\vec{k}) = \hbar v_F \, (k_x + i k_y)$$

single brand of right-handed circularly polarized photons :

$$\vec{e}_R = \frac{1}{\sqrt{2}} (1, i)$$

$$\Rightarrow \gamma(\vec{k} - \vec{A}) \longrightarrow \hbar v_F \left(k_x + i k_y - \sqrt{2} A_0 a^+ \right)$$

Define $\quad g \equiv \hbar v_F A_0 \sqrt{2}$

$$\Rightarrow H(k) = \begin{bmatrix} 0 & \hbar v_F (k_x + i k_y) - g \, a^+ \\ \hbar v_F (k_x - i k_y) - g a & 0 \end{bmatrix}$$

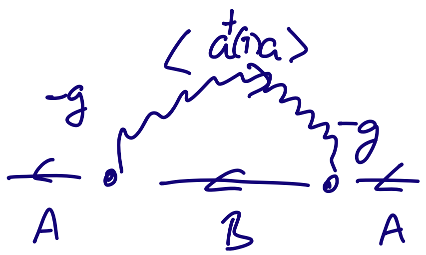
$\rightarrow$ do perturbation theory

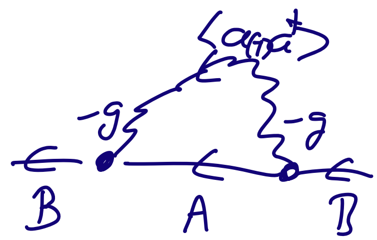
$$\hat{G}(k, \tau) = - T_\tau \begin{pmatrix} \langle C_{A,k}(\tau) C^+_{A,k} \rangle & AB \\ BA & BB \end{pmatrix}$$

$$\hat{G}^{-1} = \hat{G}_0^{-1} - \hat{\Sigma}_0$$

$$\Sigma_{0,aa}(\vec{k}, ip_n) = -\frac{g^2}{\beta}\sum_m \frac{-1}{i\omega_m+\omega}\, G_{0,bb}(\vec{k}, ip_n + i\omega_m)$$

$$\Sigma_{0,bb}(\vec{k}, ip_n) = -\frac{g^2}{\beta}\sum_m \frac{1}{i\omega_m-\omega}\, G_{0,aa}(\vec{k}, ip_n + i\omega_m)$$





$-g\, a^\dagger\, c^\dagger_{A,k}\, c_{B,k}$

$-g\, a\, c^\dagger_{B,k}\, c_{A,k}$

and $\quad G_{0,aa}(\vec{k}, ip_n) = G_{0,bb}(\vec{k}, ip_n) = \dfrac{-ip_n}{p_n^2 + v_F^2 k^2}$

→ evaluate Matsubara sum by contour integral (cf. Mahan)

e.g. $\quad \Sigma_{0,aa} = -g^2 S$

$$S = \frac{1}{\beta}\sum_m \frac{1}{i\omega_m+\omega}\,\frac{(ip_n + i\omega_m)}{(p_n+\omega_m)^2 + v_F^2 k^2}$$

$$= -\frac{1}{\beta}\sum_m f(i\omega_m)$$

use integral $I = \lim_{R\to\infty} \oint \frac{dz}{2\pi i} f(z)\, n_B(z)$

$$f(z) = \frac{1}{z+\omega} \frac{ip_n + z}{(ip_n+z)^2 - v_F^2 k^2}$$

[...]

Final result at Dirac point after analytical continuation $ip_n \to \epsilon + i0^+$:

$$\Sigma^R_{0,aa}(\vec{k}=0,\varepsilon) = \frac{g^2}{2} \frac{1}{\varepsilon + i0^+ - \omega}$$

$$\Sigma^R_{0,bb}(\vec{k}=0,\varepsilon) = \frac{g^2}{2} \frac{1}{\varepsilon + i0^+ + \omega}$$

$$\Rightarrow \text{gap } \Delta = \frac{g^2}{\omega} \underset{\underset{\frac{2g^2}{\omega^2} \ll 1}{\uparrow}}{\approx} \frac{2 v_F^2 A_0^2}{\omega} \quad \left(\begin{matrix} e \equiv \hbar \\ \equiv 1 \end{matrix} \right)$$

$\rightarrow$ same form as in Floquet case!

only difference: A_0 is not the amplitude of classical field, but of quantum fluctuations

Summary:

- dynamics after pump excitation often "O.K." described by kinetic equations (two-temperature model), but they fail to describe more interesting phenomena:
  - short-time (few fs) dynamics
  - ordered states
  - Floquet photodressing
- Floquet engineering useful to modify effective Hamiltonians (but issues in materials: heating, need for strong lasers, short-lived effects only during pump)
- Cavity engineering can be used to achieve similar effects as in Floquet but in cavity-matter ground state (no heating, long-lived); but need to engineer small effective mode volumes, near-field effects, strong light-matter coupling