Light-induced states of matter
from Floquet engineering to cavity materials

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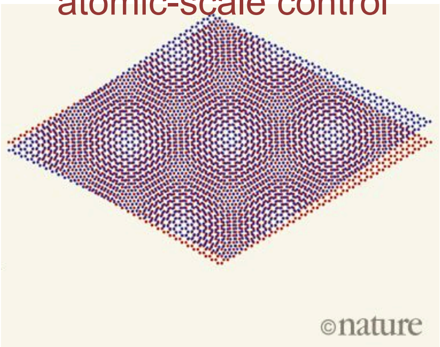
Max Planck Institute for the Structure and Dynamics of Matter

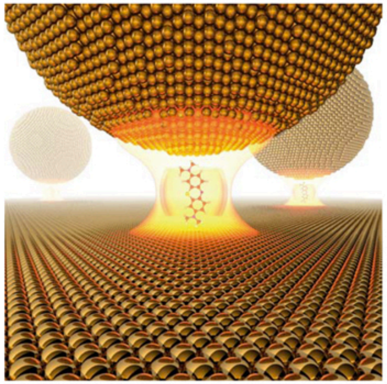
Engineering materials with light

condensed matter
quantum materials
atomic-scale control



Y. Cao et al., Nature 556, 43 (2018)

nonequilibrium materials engineering



R. Chikkaraddy et al., Nature 535, 127 (2016)

quantum optics
nanoplasmonics
polaritonic chemistry
QED: vacuum fluctuations

ultrafast spectroscopy
revealing elementary couplings
light-induced new states of matter

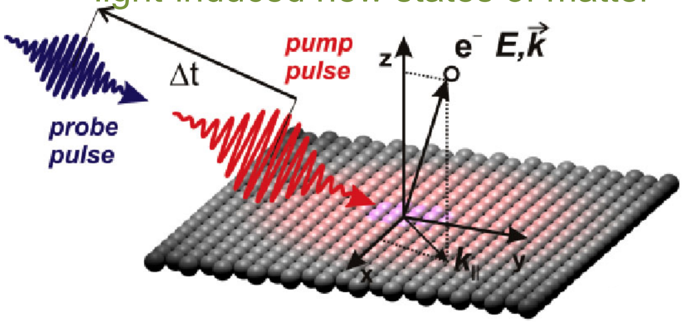


Image courtesy: J. Sobota

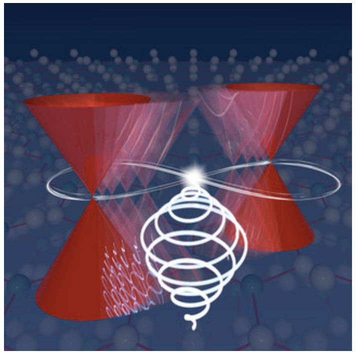
pump-probe: strong classical fields

Outline

1. Floquet engineering

   coherent laser driving can induce topology

   M. A. Sentef et al., Nat. Commun. 6, 7047 (2015)
   H. Hübener et al., Nat. Commun. 8, 13940 (2017)
   G. E. Topp et al., PRResearch 1, 023031 (2019)



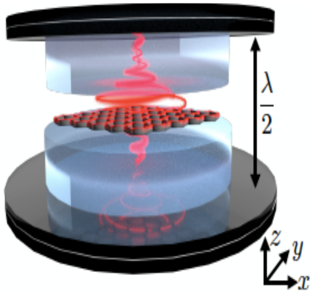
2. Cavity engineering

   light-induced topology from pure vacuum fluctuations of light

   X. Wang, E. Ronca, M. A. Sentef, PRB 99, 235156 (2019)

   cavity superconductivity

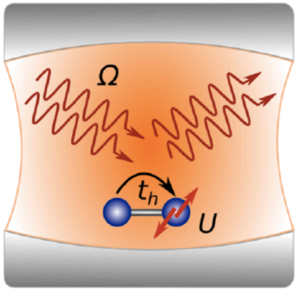
   M. A. Sentef, M. Ruggenthaler, A. Rubio, Science Advances 4, eaau6969 (2018)



3. Cavity to Floquet crossover

   strong light-matter coupling: Floquet effects without coherence

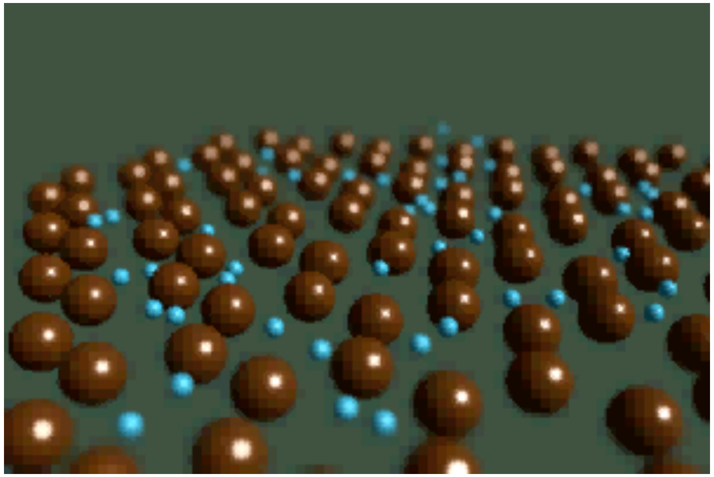
   M. A. Sentef, J. Li, F. Künzel, M. Eckstein, PRResearch 2, 033033 (2020)
   E. V. Boström, M. Claassen, J. W. McIver, G. Jotzu, A. Rubio, M. A. Sentef, arXiv: 2007.01714



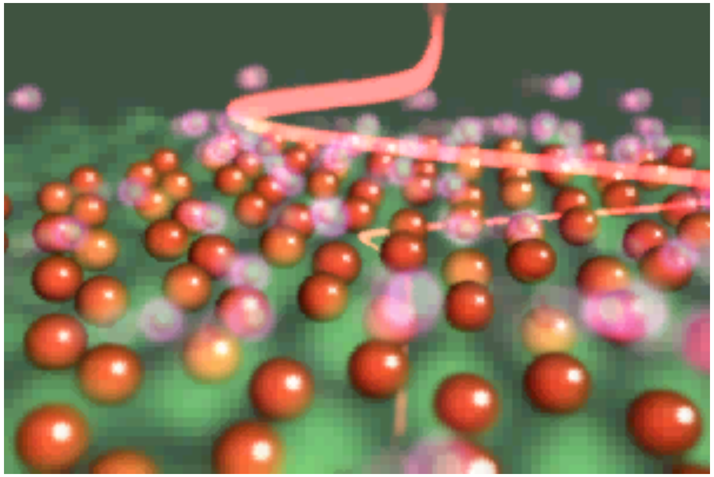
1. Floquet engineering

Floquet states of matter

by Koichiro Tanaka (Kyoto university)



electrons in solids



Floquet state (photo-dressed state)

$$H$$

$$H_{\text{eff}}$$

$$H_{\text{eff}} = H_0 + \frac{[H_{-1}, H_1]}{\Omega} + \mathcal{O}(\Omega^{-2})$$

Floquet states of matter

mpsd

time periodic system

$$i\partial_t\psi = H(t)\psi \qquad H(t) = H(t+T) \quad \Omega = 2\pi/T$$

"Floquet mapping"

=discrete Fourier trans.

$$\Psi(t) = e^{-i\varepsilon t}\sum_m \phi^m e^{-im\Omega t}$$

Floquet Hamiltonian (static eigenvalue problem)

$$\sum_{m=-\infty}^{\infty} \mathcal{H}^{mn}\phi_\alpha^m = \varepsilon_\alpha \phi_\alpha^n$$

ε: Floquet quasi-energy

$$(\mathcal{H})^{mn} = \frac{1}{T}\int_0^T dt H(t) e^{i(m-n)\Omega t} + m\delta_{mn}\Omega I$$

comes from the $i\partial_t$ term

$$H_m = \mathcal{H}^{m0}$$

~ absorption of m "photons"

Floquet states of matter

Time-periodic quantum system = Floquet theory (exact) ~ effective theory

$$i\partial_t \psi = H(t)\psi$$

$$H(t) = H(t+T)$$

$$\mathcal{H}\phi = \varepsilon\phi$$

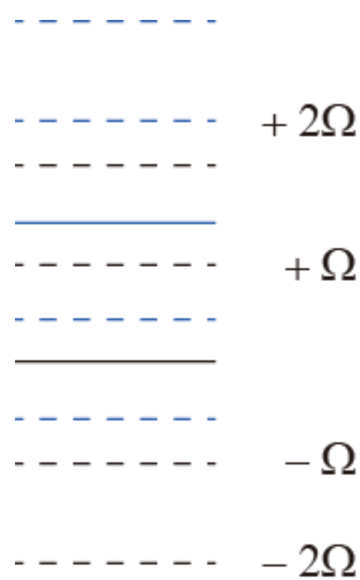
$$H_{\text{eff}} = H_0 + \frac{[H_{-1}, H_1]}{\Omega} + \mathcal{O}(\Omega^{-2})$$

Fictitious fields!

Floquet theory

projection to the original Hilbert space

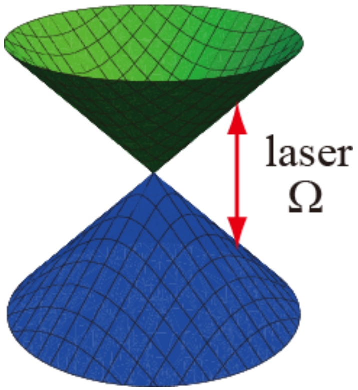
two states + periodic driving



Hilbert space size
= original system

n-photon dressed state
Floquet side bands

Dirac fermion + circularly polarized laser



coupling to AC field

$$\boldsymbol{k} \rightarrow \boldsymbol{k} + \boldsymbol{A}(t)$$

$$k = k_x + ik_y$$
$$\boldsymbol{A}(t) = (F/\Omega \cos \Omega t,\, F/\Omega \sin \Omega t)$$
$$A = F/\Omega$$

time dependent Schrödinger equation

$$i\partial_t \psi_k = \begin{pmatrix} 0 & k + Ae^{i\Omega t} \\ \bar{k} + Ae^{-i\Omega t} & 0 \end{pmatrix} \psi_k$$

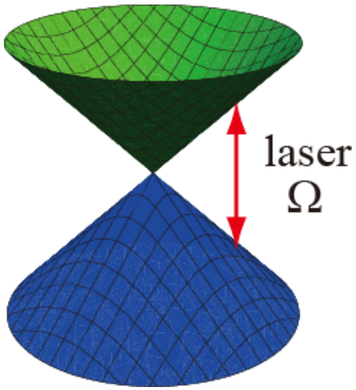
Floquet theory

$$(\mathcal{H})^{mn} = \frac{1}{T}\int_0^T dt H(t) e^{i(m-n)\Omega t} + m\delta_{mn}\Omega I$$

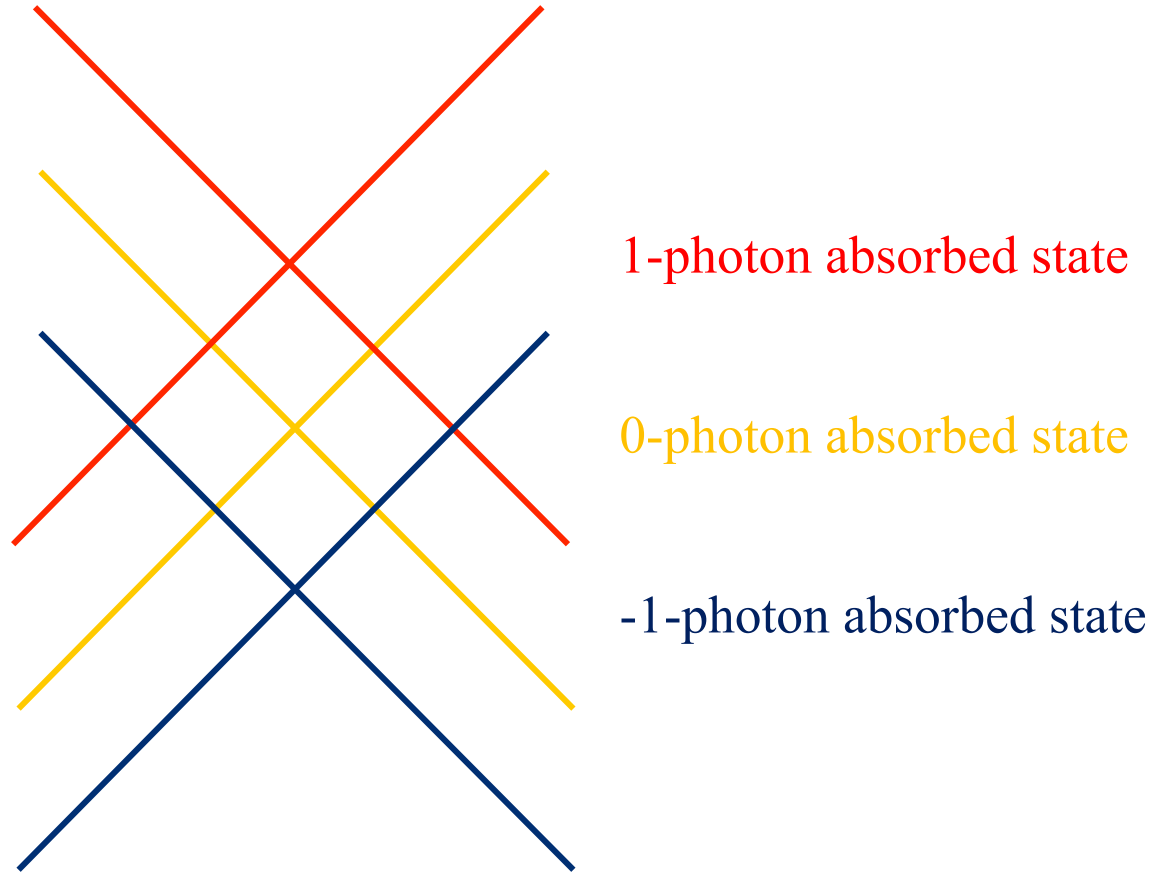
$$H^{\text{Floquet}} = \begin{pmatrix} \Omega & k & 0 & A & 0 & 0 \\ \bar{k} & \Omega & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & k & 0 & A \\ A & 0 & \bar{k} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -\Omega & k \\ 0 & 0 & A & 0 & \bar{k} & -\Omega \end{pmatrix}$$

truncated at m=0,+1, -1 for display

Dirac fermion + circularly polarized laser



$$H^{\text{Floquet}} = \begin{pmatrix} \Omega & k & 0 & A & 0 & 0 \\ \bar{k} & \Omega & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & k & 0 & A \\ A & 0 & \bar{k} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -\Omega & k \\ 0 & 0 & A & 0 & \bar{k} & -\Omega \end{pmatrix}$$

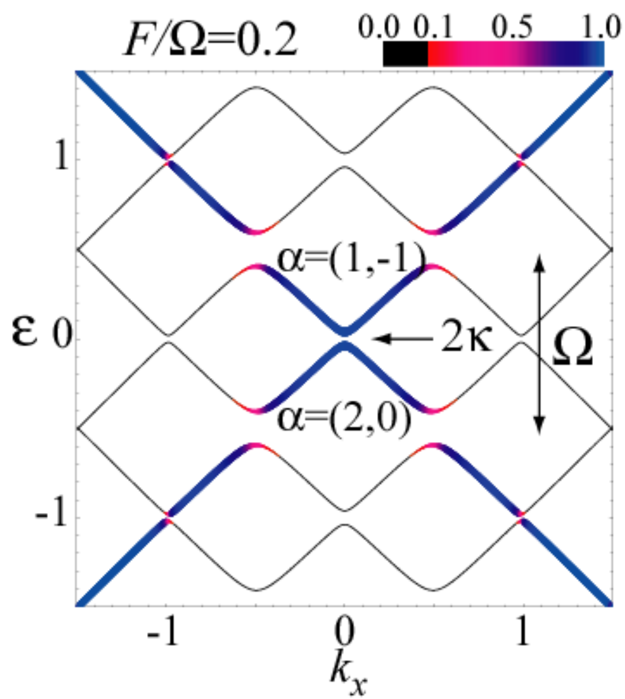


Dirac fermion + circularly polarized laser

Mass term =
synthetic field stemming from a real time-dependent field A(t)

$$\kappa = \frac{\sqrt{4A^2+\Omega^2}-\Omega}{2} \sim A^2/\Omega$$

$$H^{\text{Floquet}} = \begin{pmatrix} \Omega & k & 0 & A & 0 & 0 \\ \bar{k} & \Omega & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & k & 0 & A \\ A & 0 & \bar{k} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -\Omega & k \\ 0 & 0 & A & 0 & \bar{k} & -\Omega \end{pmatrix}$$



1-photon absorbed state

0-photon absorbed state

-1-photon absorbed state

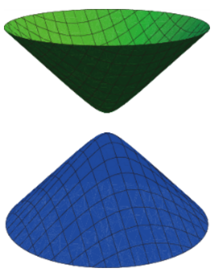
Oka and Aoki, PRB 79, 081406 (2009)

Dirac fermion + circularly polarized laser

Projection to the original Hilbert space

$$H^{\mathrm{Floquet}} = \begin{pmatrix} \Omega & k & 0 & A & 0 & 0 \\ \bar{k} & \Omega & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & k & 0 & A \\ A & 0 & \bar{k} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -\Omega & k \\ 0 & 0 & A & 0 & \bar{k} & -\Omega \end{pmatrix}$$

near Dirac point



2κ

Dynamical gap

$$\kappa = \frac{\sqrt{4A^2+\Omega^2}-\Omega}{2} \sim A^2/\Omega$$

2nd order perturbation

$$H_{\mathrm{eff}} = H_0 + \frac{[\overset{\sim A\sigma_-}{H_{-1}}, \overset{\sim A\sigma_+}{H_1}]}{\Omega} + \mathcal{O}(A^4)$$

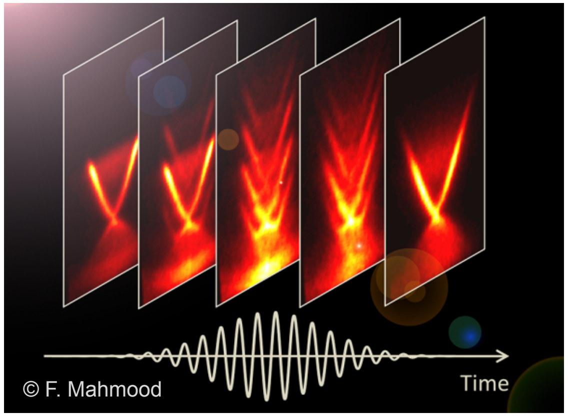
Mass term =
synthetic field stemming from a
real time-dependent field A(t)

$$\sim v(k_x\sigma_y - \tau_z k_y \sigma_x) \pm \tau_z \frac{v^2 A^2}{\Omega}\sigma_z$$

$$A = F/\Omega$$

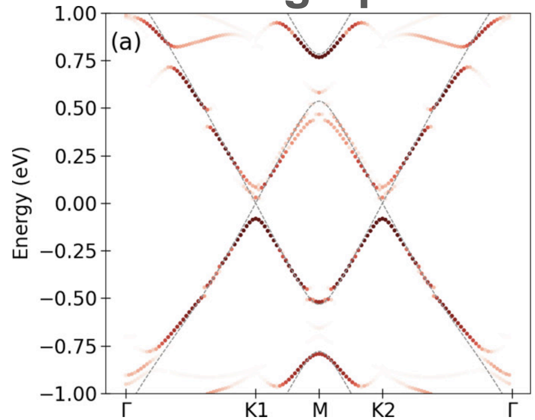
Floquet at work

Floquet-Bloch bands in time-resolved ARPES



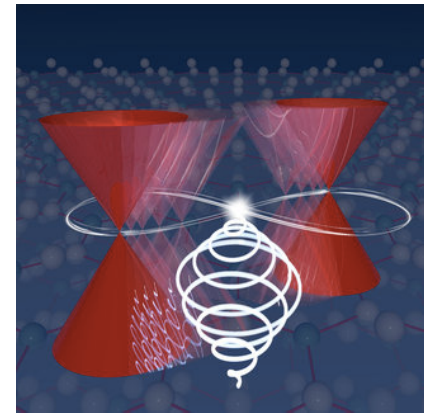
F. Mahmood et al., Nature Physics 12, 306 (2016)

Floquet topology in Moiré graphene

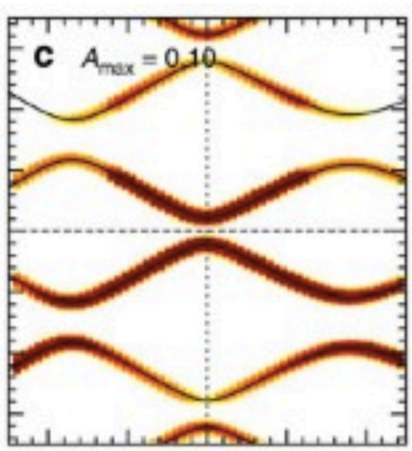


G. E. Topp et al., PRResearch 1, 023031 (2019)

Floquet-Weyl semimetal



H. Hübener et al., Nat. Commun. 8, 13940 (2017)



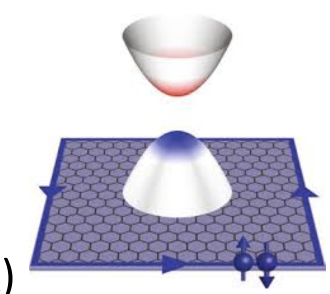
M. A. Sentef et al., Nat. Commun. 6, 7047 (2015)

... but many more theory Floquet proposals than experiments in materials. Issues:

- need for strong lasers
- need for coherence
- detrimental heating effects

possible resolution: cavities (next part of talk)

Light-induced anomalous Hall effect



James McIver's talk

2. Cavity engineering

Cavity QED matter coupling

mpsd

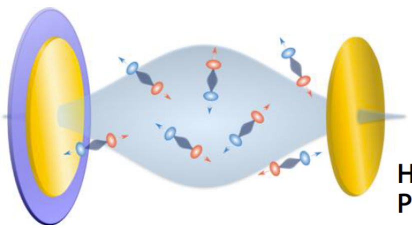
CAVITY QUANTUM ELECTRODYNAMICS

A new generation of experiments shows that spontaneous radiation from excited atoms can be greatly suppressed or enhanced by placing the atoms between mirrors or in cavities.

Serge Haroche and Daniel Kleppner   Physics Today 1989

changing the vacuum changes the matter!

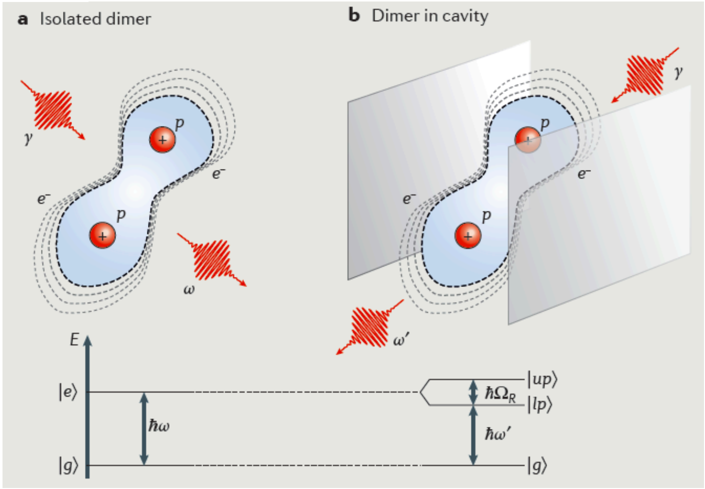
Recent years: Placing atoms and molecules in cavities shown to sometimes dramatically change their properties and chemical reactions. Scientists talk about „light-matter (collective) strong coupling“.



Hybrid Light–Matter States in a Molecular and Material Science Perspective

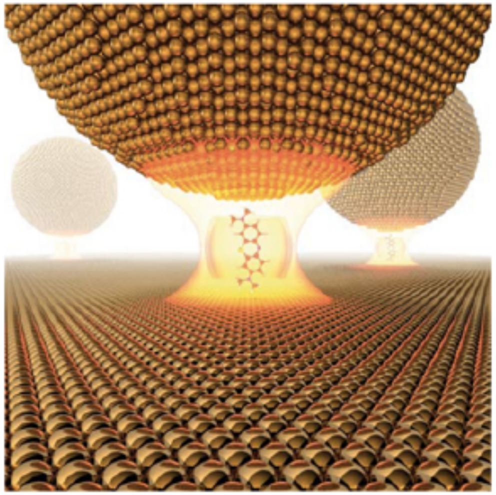
T. Ebbesen, Acc. Chem. Res. 49, 2403 (2016)

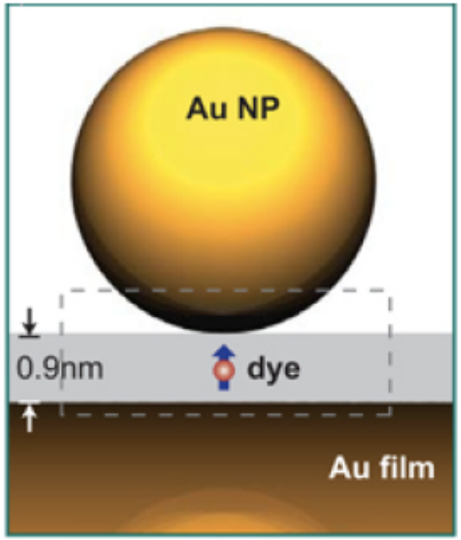
higher enhancements. Another direction is to check physical phenomena that are sensitive to phonon energy. Metal–insulating and superconducting transitions for instance might be significantly modified under strong coupling.



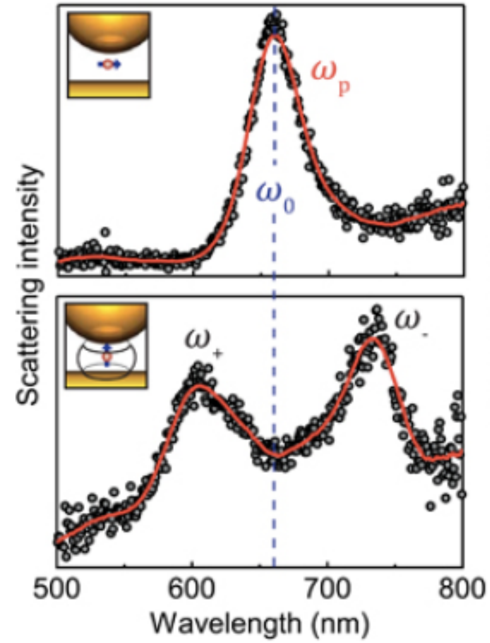
M. Ruggenthaler et al., Nat. Rev. Chem. 2, 0118 (2018)
J. Feist et al., ACS Photonics 5, 205 (2017)
R. F. Ribeiro et al., Chem. Sci. 9, 6325 (2018)
J. Flick et al., Nanophotonics 7, 1479 (2018)
A. F. Kockum et al., Nat. Rev. Phys. 1, 19 (2019)

From classical to quantum light

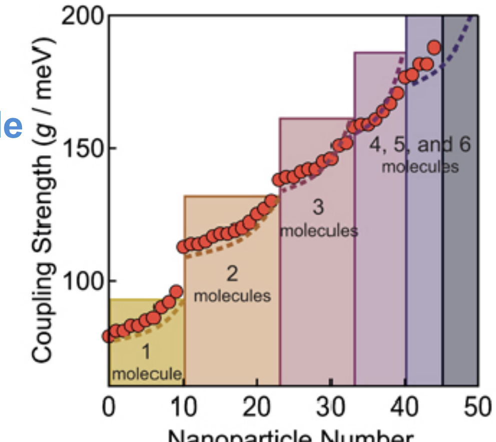




R. Chikkaraddy et al., Nature 535, 127 (2016)



Rabi splitting



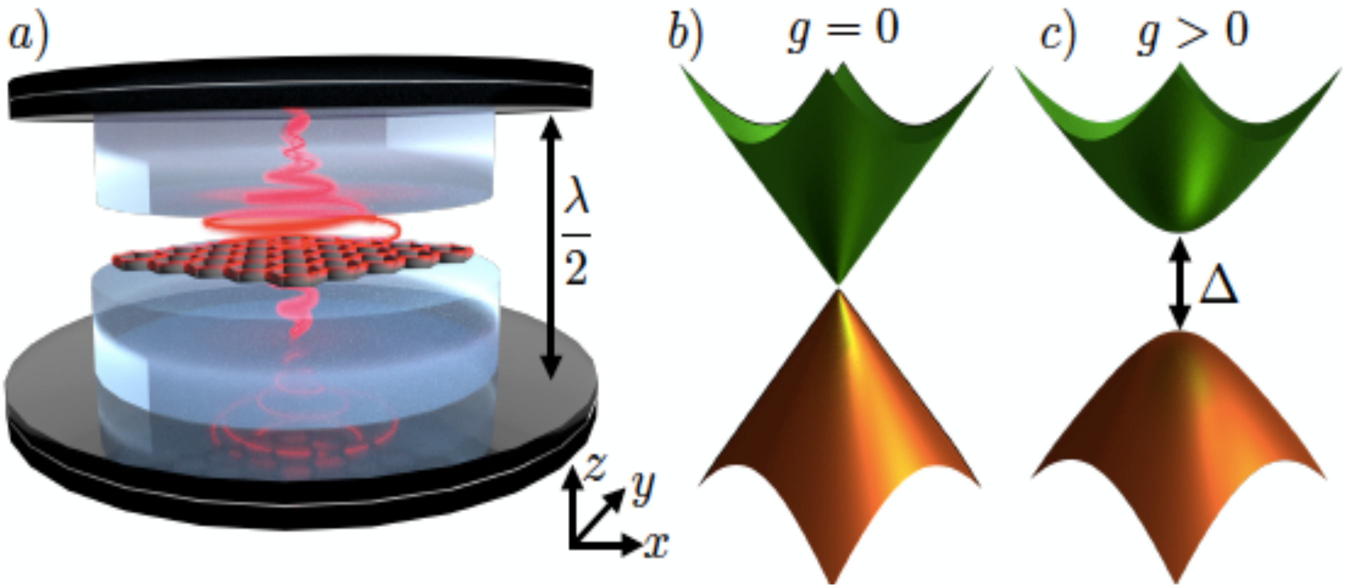
collective strong light-matter coupling
when many atoms interact with the same cavity photon mode

cavity materials: many atoms interact with the same modes

Cavity-induced topology

Cavity-induced quantized anomalous Hall effect in graphene

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



Dirac fermion in cavity

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

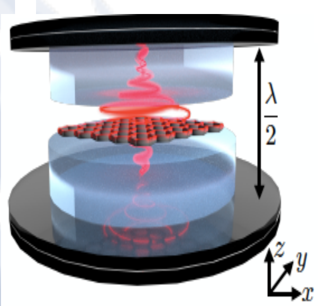
Dirac cone couples to cavity modes: $\gamma(\vec{k}-\hat{\vec{A}}) \rightarrow \hbar v_F(k_x + ik_y - \sqrt{2}A_0 a^\dagger)$

$$H = \sum_{\vec{k}} \begin{pmatrix} c^\dagger_{A,\vec{k}} \\ c^\dagger_{B,\vec{k}} \end{pmatrix}^T \begin{pmatrix} 0 & \gamma(\vec{k}-\hat{\vec{A}}) \\ \gamma(\vec{k}-\hat{\vec{A}})^\dagger & 0 \end{pmatrix} \begin{pmatrix} c_{A,\vec{k}} \\ c_{B,\vec{k}} \end{pmatrix} + \sum_\lambda \omega_\lambda a^\dagger_\lambda a_\lambda ,$$

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

$$A_0 = \sqrt{\hbar/(\epsilon\epsilon_0 V\omega)}$$

cavity coupling controlled by mode volume V, dielectric environment ε, and mode frequency ω



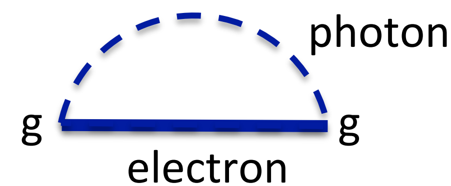
exchange of virtual photons with the cavity vacuum

Dirac fermion in cavity

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

Using a right-handed circularly polarized cavity reduces the photon field to a single branch with $\vec{e}_\lambda \equiv \vec{e}$, operators $a_\lambda^\dagger \equiv a^\dagger$, and frequency $\omega_\lambda \equiv \omega$, with unit polarization vector $\vec{e} = \frac{1}{\sqrt{2}}(1, i)$. In this case, $\gamma(\vec{k} - \hat{\vec{A}}) \rightarrow \hbar v_F (k_x + i k_y - \sqrt{2} A_0 a^\dagger)$

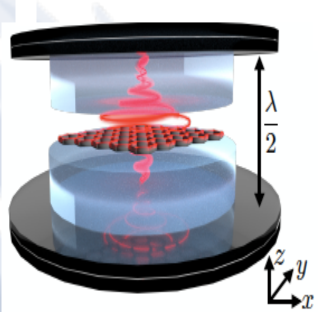
band renormalization due to electron-photon self-energy



$$g \equiv v_F A_0 \sqrt{2}$$

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

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



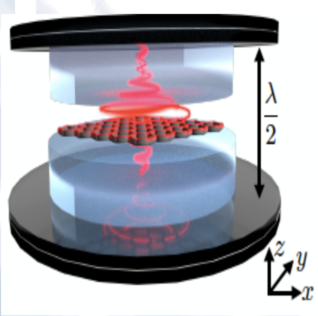
Dirac fermion in cavity

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

Energy gap at Dirac point: $\Delta = \sqrt{2g^2 + \omega^2} - \omega$

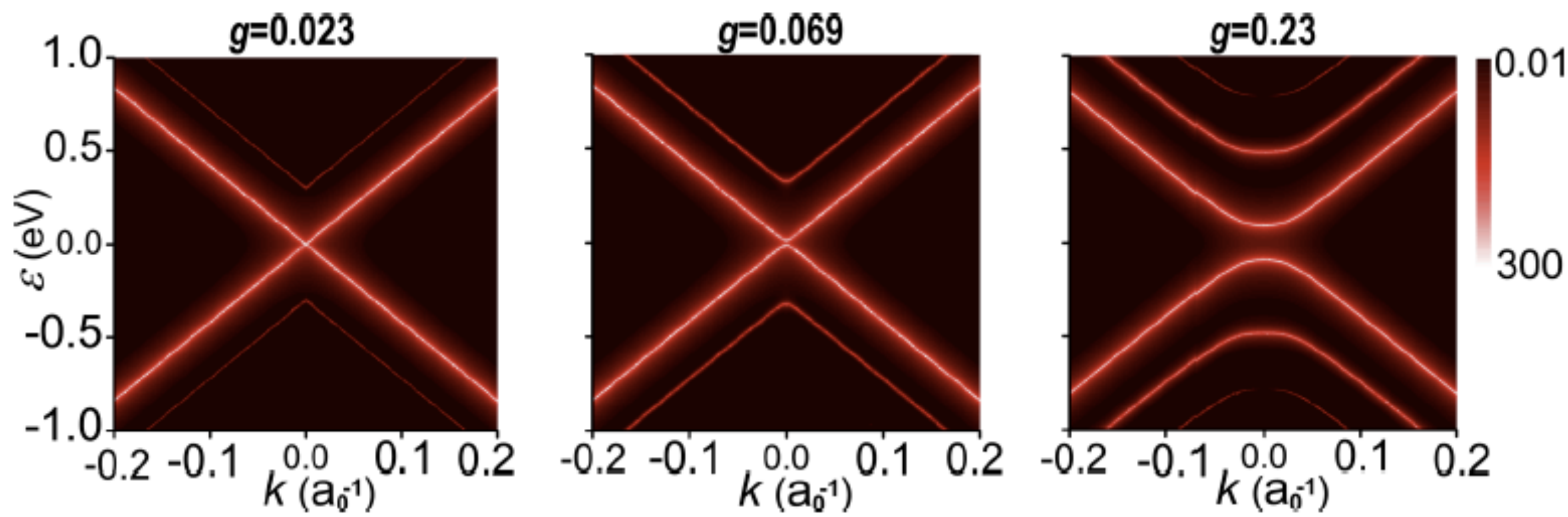
In the limit $2g^2/\omega^2 \ll 1$, we obtain $\Delta \approx \frac{g^2}{\omega} = \frac{2\hbar^2 v_F^2 A_0^2}{\omega}$

... looks like Floquet result but different interpretation of A_0:
- Floquet = classical limit: A_0 is the laser field amplitude
- dark cavity = quantum limit: A_0 is the amplitude of quantum fluctuations



Dirac fermion in cavity

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

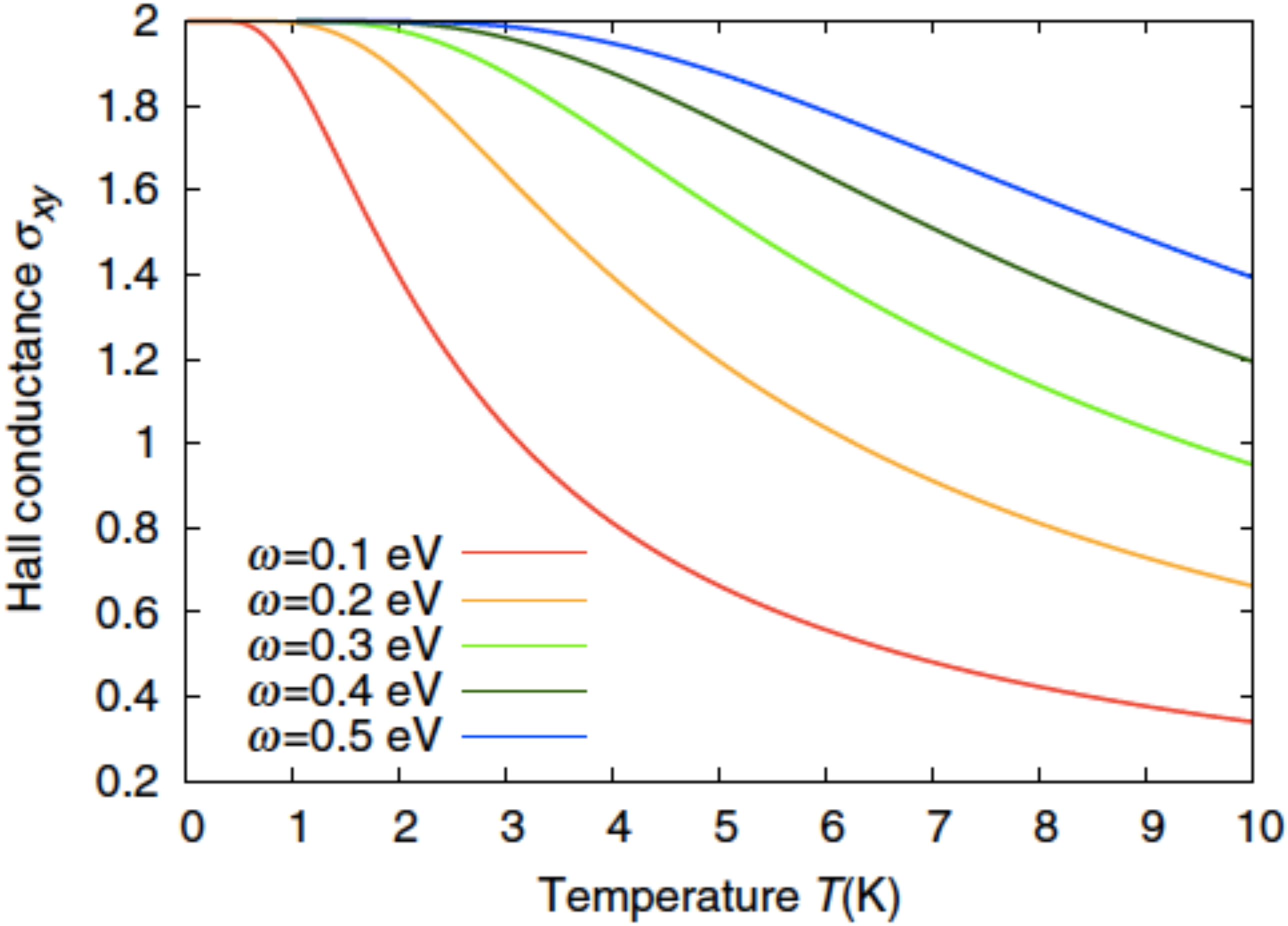


energy gap and photon sidebands, controlled by light-matter coupling strength

Dirac fermion in cavity

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

mpsd

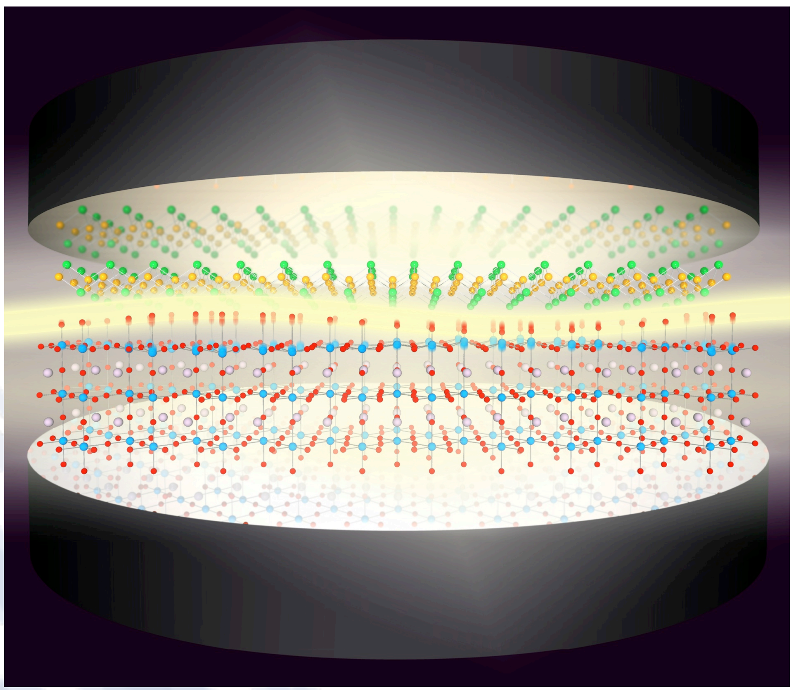


quantized light-induced Hall conductance at low temperatures, controlled by cavity geometry

Cavity superconductivity?

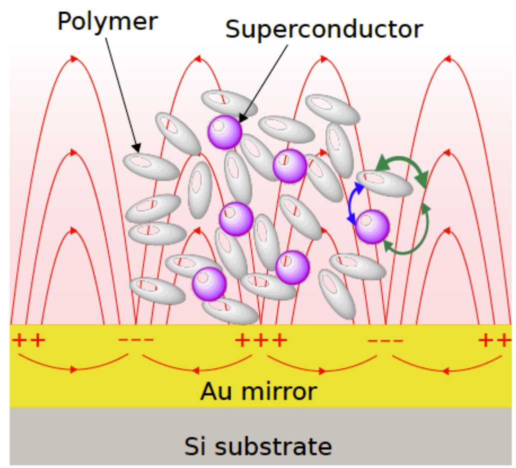
Cavity quantum-electrodynamical polaritonically enhanced electron-phonon coupling and its influence on superconductivity

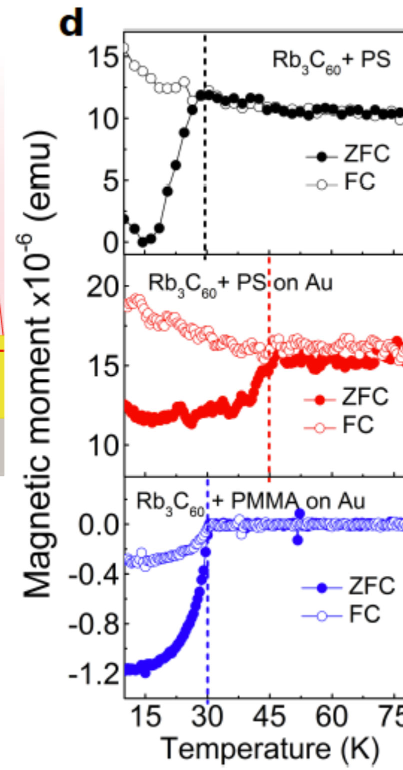
M. A. Sentef, M. Ruggenthaler, A. Rubio, Science Advances 4, eaau6969 (2018)



Exploring Superconductivity under Strong Coupling with the Vacuum Electromagnetic Field arXiv:1911.01459

A. Thomas[1], E. Devaux[1], K. Nagarajan[1], T. Chervy[1], M. Seidel[1], D. Hagenmüller[1], S. Schütz[1], J. Schachenmayer[1], C. Genet[1], G. Pupillo[1*] & T. W. Ebbesen[1*]





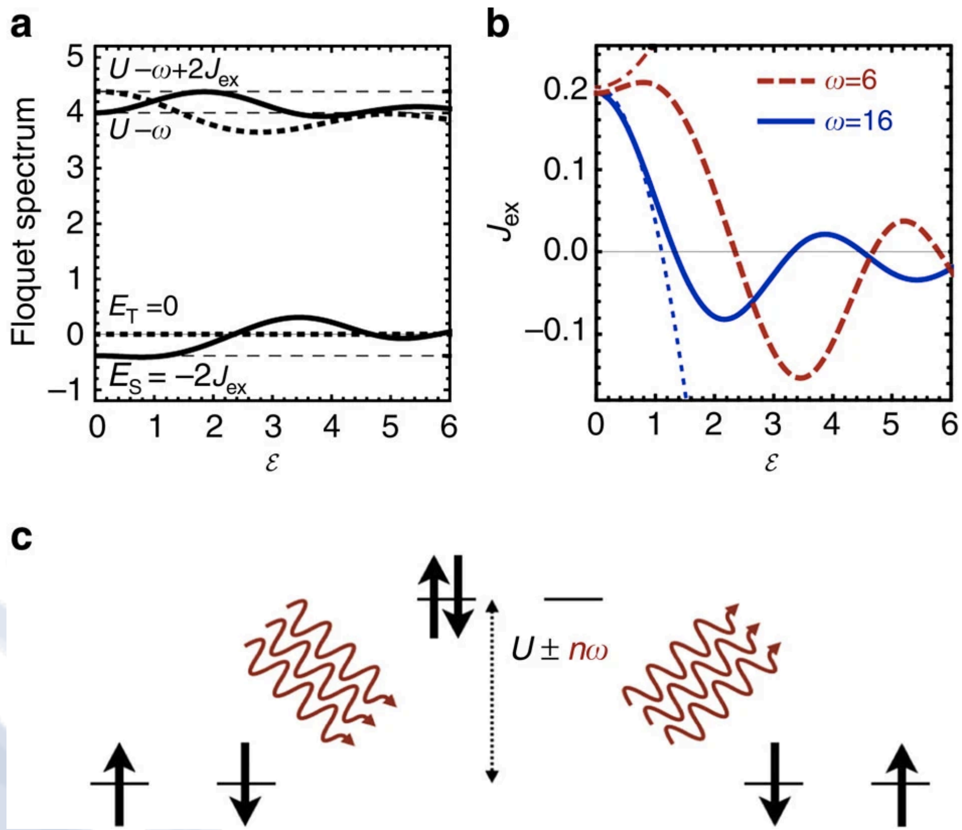
suggests enhanced electron-phonon coupling due to polariton formation and mode softening

3. Cavity to Floquet crossover

Cavity to Floquet crossover

M. A. Sentef, J. Li, F. Künzel, M. Eckstein, PRResearch 2, 033033 (2020)

Motivation: Ultrafast and reversible control of exchange interaction with classical field
Mentink, Balzer, and Eckstein, Nat. Commun. 6, 6708 (2015)



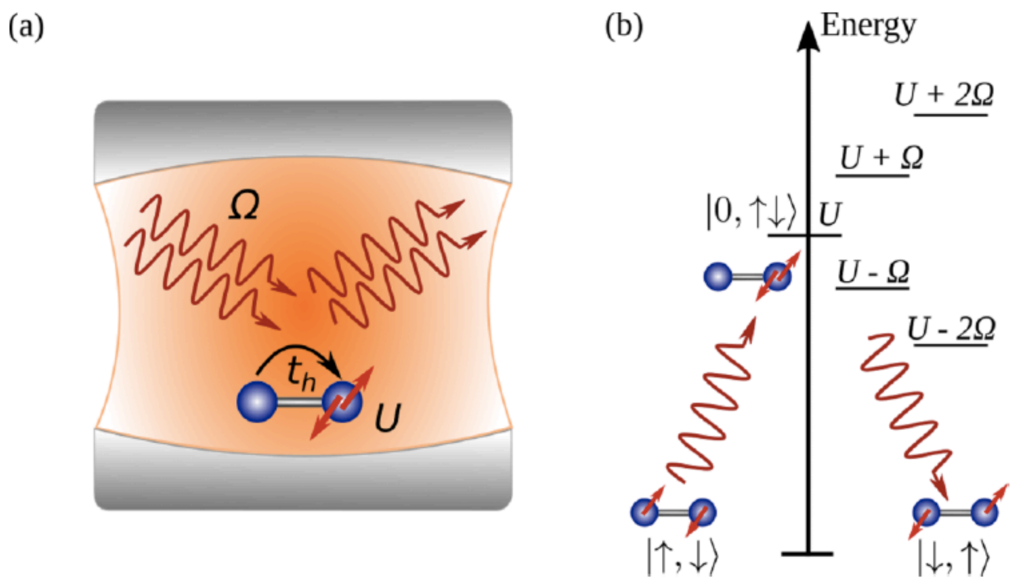
emission and absorption of real photons during exchange process renormalizes J_{ex}

sign of change of J_{ex} controlled by detuning of laser frequency from Hubbard U at small field strength

what about the cavity limit?
can we investigate the crossover?

Cavity to Floquet crossover

M. A. Sentef, J. Li, F. Künzel, M. Eckstein, PRResearch 2, 033033 (2020)



$$\hat{H} = t_h \sum_{j\sigma} (\hat{c}^\dagger_{j,\sigma} \hat{c}_{j+1,\sigma}\, e^{i\hat{A}} + \text{H.c.}) + U \sum_j \hat{n}_{j,\uparrow} \hat{n}_{j,\downarrow} + \Omega \hat{a}^\dagger \hat{a}.$$

$$\hat{A} = g(\hat{a} + \hat{a}^\dagger)$$

A: effective vector potential

g: light-matter coupling strength

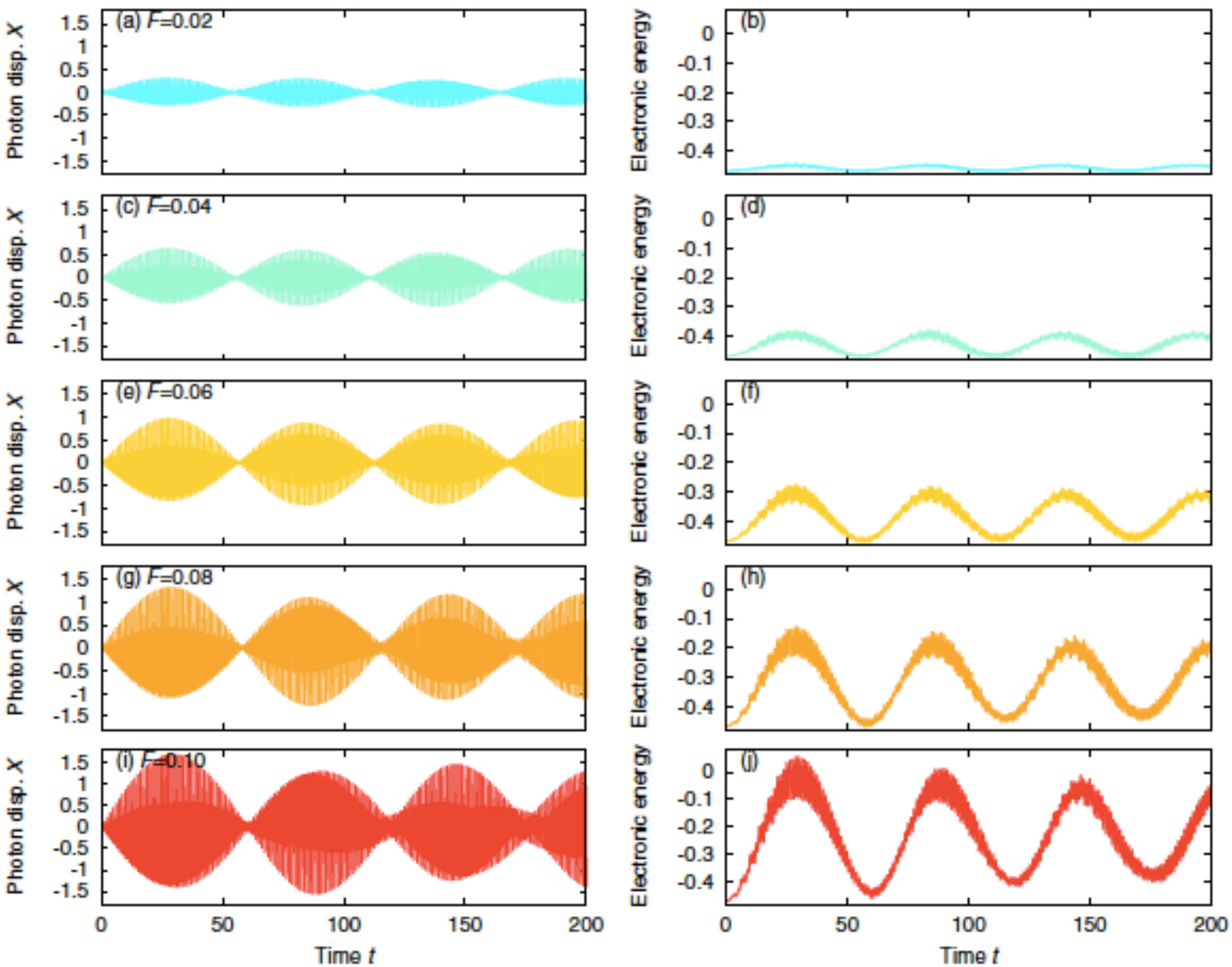
Quantum system -> Floquet system for $n \to \infty, \quad g\sqrt{n}$ fixed.

(large photon number, weak light-matter coupling strength g)

Photon number states are good enough to see Floquet-engineering effects at sufficiently large coupling strength – coherent states not required!

Driven cavity

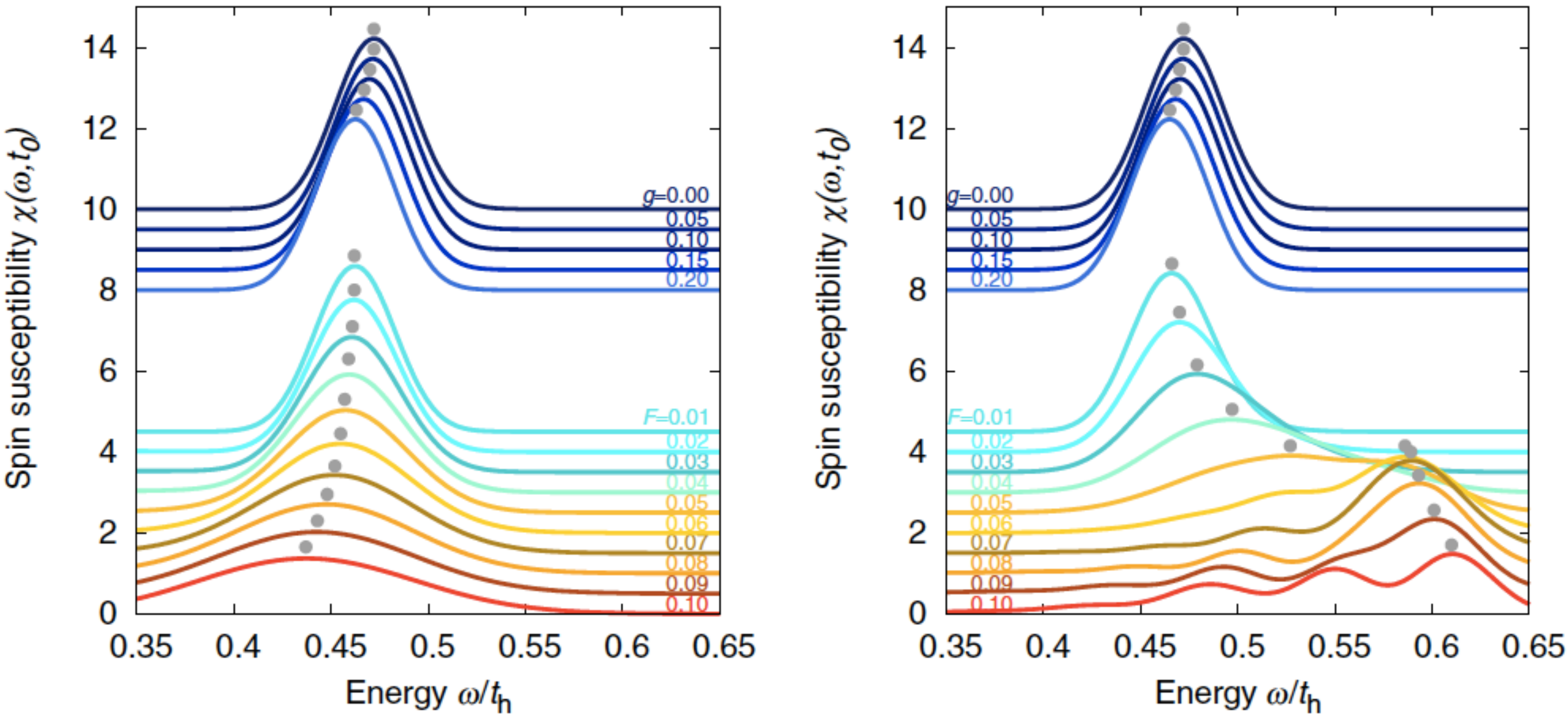
M. A. Sentef, J. Li, F. Künzel, M. Eckstein, PRResearch 2, 033033 (2020)



Driven cavity

M. A. Sentef, J. Li, F. Künzel, M. Eckstein, PRResearch 2, 033033 (2020)

Time-resolved spin susceptibility in resonantly driven cavity (peak position ~ J_{ex})



Cavity frequency > U

Cavity frequency < U

J_{ex} always reduced by vacuum fluctuations

blue-detuned: J_{ex} further reduced by driving; red-detuned: J_{ex} enhanced by driving

Floquet engineering without macroscopic laser fields

M. A. Sentef, J. Li, F. Künzel, M. Eckstein, PRResearch 2, 033033 (2020)

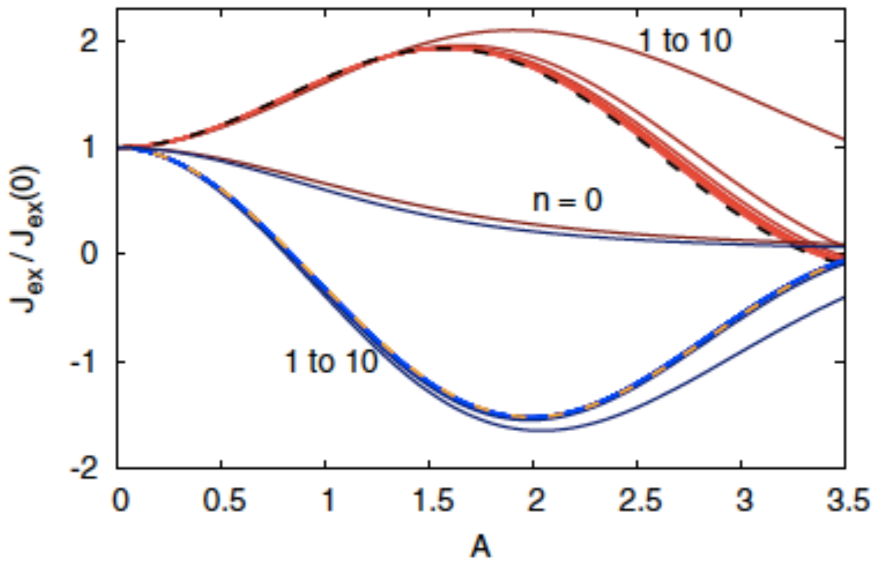


FIG. 4. Exchange interaction $J_{ex}^{(n)}$ in the n-photon state as a function of A for $n = 1, \ldots, 10$. The vertical axis is in units of $J_{ex}(0)$ at coupling $g = 0$. The curves with colors from dark to light red correspond to $\Omega = 0.8U$ (red-detuned) and those with dark to light blue correspond to $\Omega = 1.2U$ (blue-detuned). The lightness of the color indicates the photon number n with the darkest ones denoting $n = 0$. The dashed black (orange) line shows the Floquet result (11) for $\Omega = 0.8U$ ($\Omega = 1.2U$). For the dark-cavity ($n = 0$) case, $J_{ex}^{(0)}$ is plotted as a function of the coupling $g = A$.

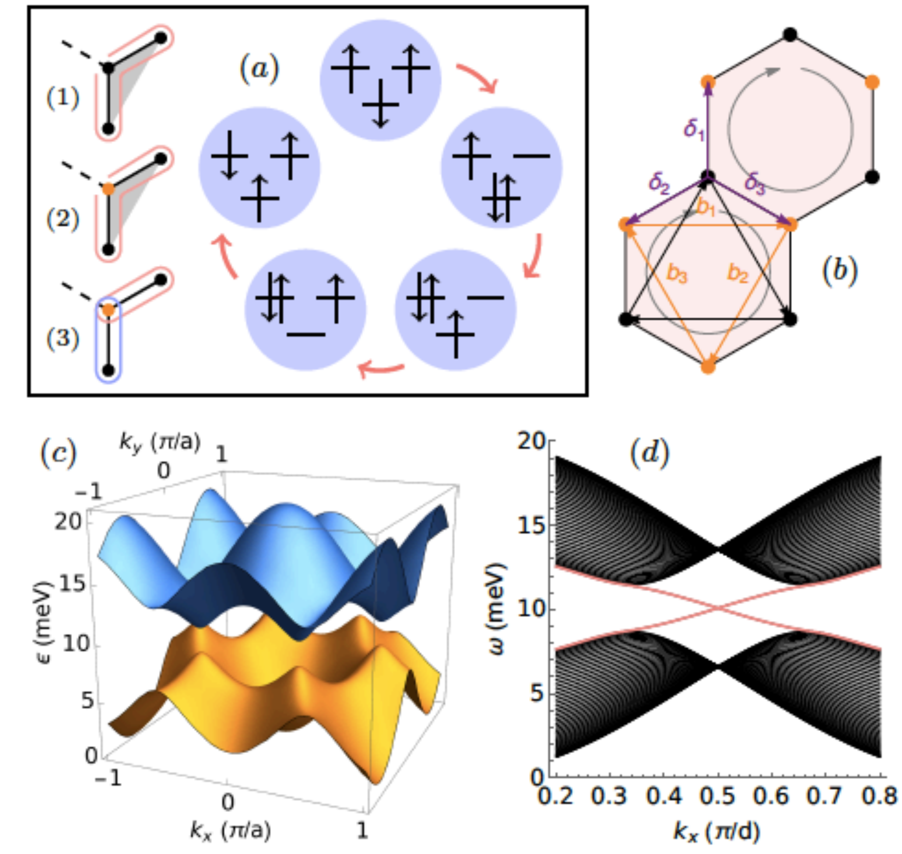
At fixed photon number,
Floquet limit is reached
as the light-matter coupling strength
is increased!

Note:
photon number states have
zero macroscopic field
-> coherence is not required at sufficiently strong coupling!

Floquet topological magnons

E. V. Boström, M. Claassen, J. W. McIver, G. Jotzu, A. Rubio, and M. A. Sentef, arXiv: 2007.01714

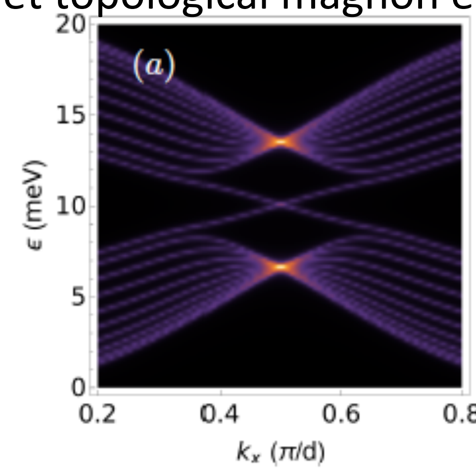
also see: Claassen et al., Nat. Commun. 8, 1192 (2017); Kitamura et al., PRB 96, 014406 (2017); Owerre, Journal of Physics Communications 1, 021002 (2017)



Light-induced scalar spin chirality in 2D honeycomb magnets

$$\mathcal{H} = \sum_{\langle ij \rangle} J_{ij} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j + \sum_{\langle\langle ik \rangle\rangle} J'_{ik} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_k + \sum_{\langle\langle ik \rangle\rangle} \chi_{ik} \hat{\mathbf{S}}_j \cdot (\hat{\mathbf{S}}_i \times \hat{\mathbf{S}}_k).$$

Floquet topological magnon edge states

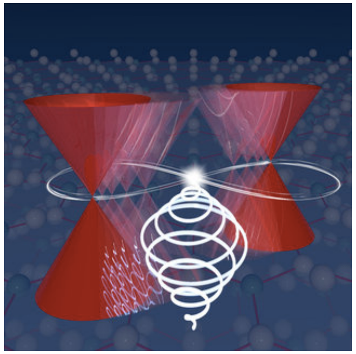


Summary

1. Floquet engineering

   coherent laser driving can induce topology

   M. A. Sentef et al., Nat. Commun. 6, 7047 (2015)
   H. Hübener et al., Nat. Commun. 8, 13940 (2017)
   G. E. Topp et al., PRResearch 1, 023031 (2019)



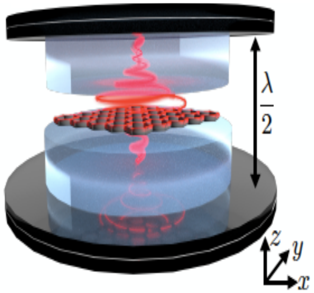
2. Cavity engineering

   light-induced topology from pure vacuum fluctuations of light

   X. Wang, E. Ronca, M. A. Sentef, PRB 99, 235156 (2019)

   cavity superconductivity

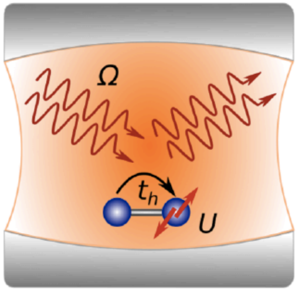
   M. A. Sentef, M. Ruggenthaler, A. Rubio, Science Advances 4, eaau6969 (2018)



3. Cavity to Floquet crossover

   strong light-matter coupling: Floquet effects without coherence

   M. A. Sentef, J. Li, F. Künzel, M. Eckstein, PRResearch 2, 033033 (2020)
   E. V. Boström, M. Claassen, J. W. McIver, G. Jotzu, A. Rubio, M. A. Sentef, arXiv: 2007.01714



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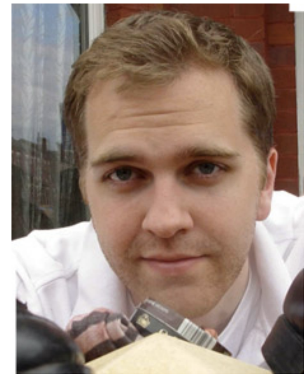
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Thank you for your kind attention!