

Engineering emergent states in quantum materials with classical and quantum light

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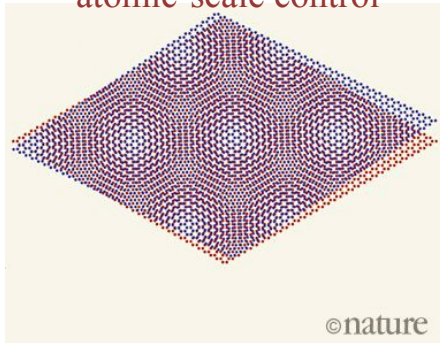
Max-Planck Institute for the Structure and Dynamics of Matter, Hamburg

DPG Surface Science Meeting, March 3, 2021

Engineering quantum materials with light

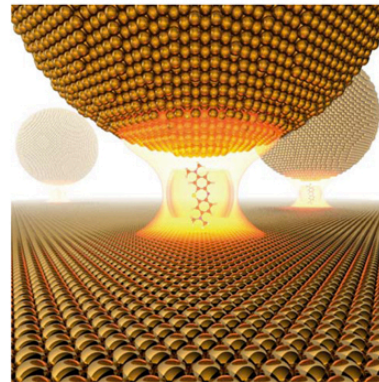
condensed matter

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

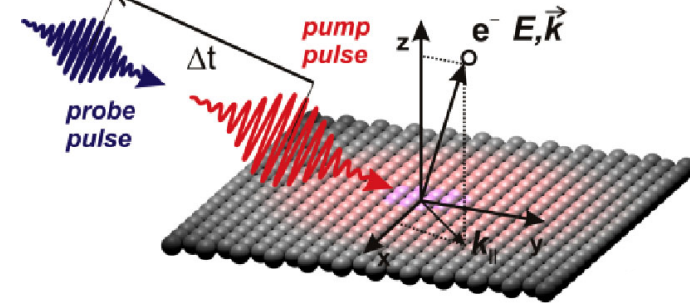


Image courtesy: J. Sobota

pump-probe: strong classical fields

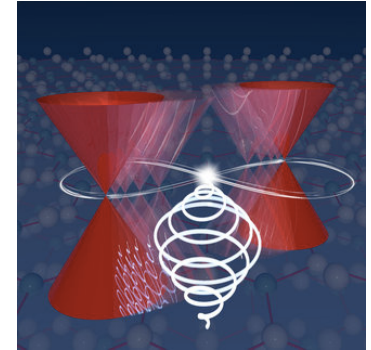
Floquet engineering → c.f. Netanel Lindner's talk

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)



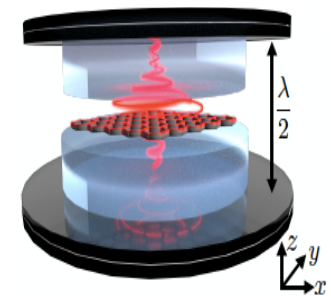
Cavity engineering

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)

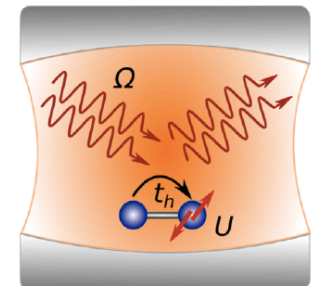


Cavity to Floquet crossover

strong light-matter coupling: Floquet effects without coherence

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

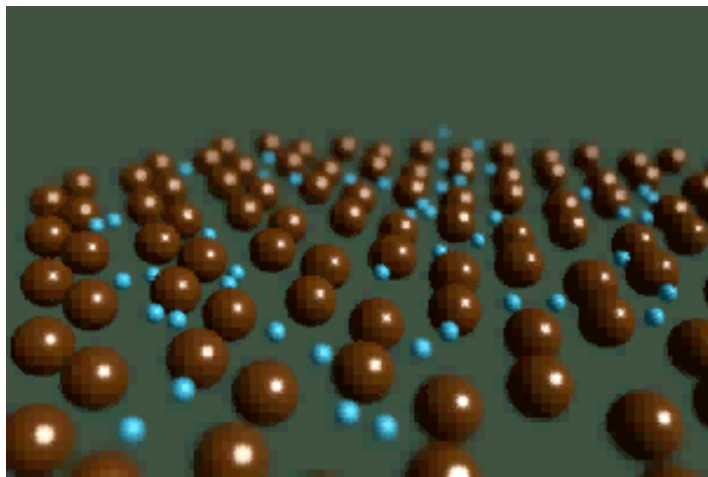
E. V. Boström, M. Claassen, J. W. McIver, G. Jotzu, A. Rubio, M. A. Sentef, arXiv: 2007.01714, SciPost Physics (2020)



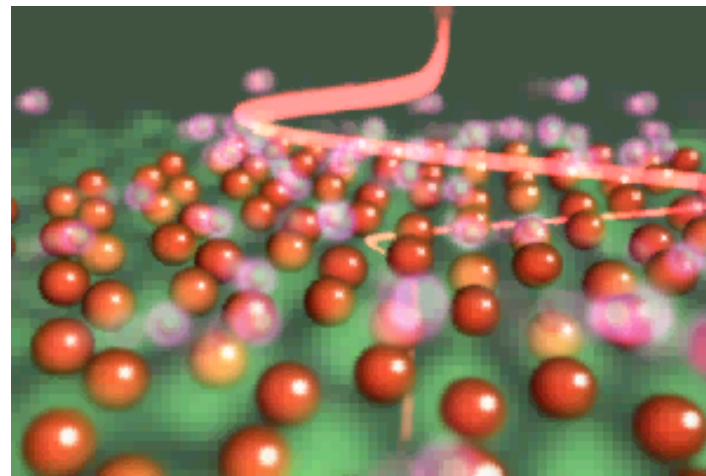
1. Floquet engineering

Floquet states of matter

by Koichiro Tanaka (Kyoto university)



electrons in solids



Floquet state (photo-dressed state)

Why Floquet ?

→ engineer
effective

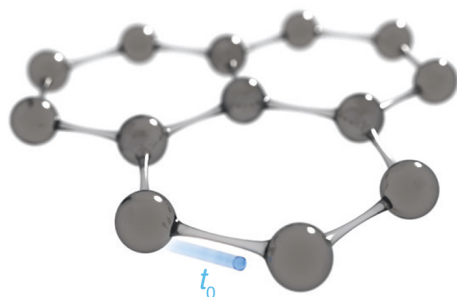
Hamiltonians

H_{eff}

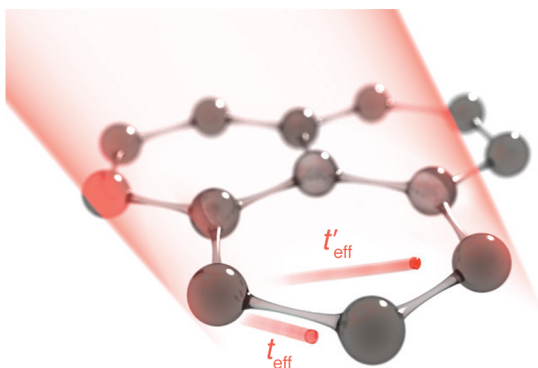
$$H_{\text{eff}} = H_0 + \frac{[H_{-1}, H_1]}{\Omega} + \mathcal{O}(\Omega^{-2})$$

Floquet states of matter

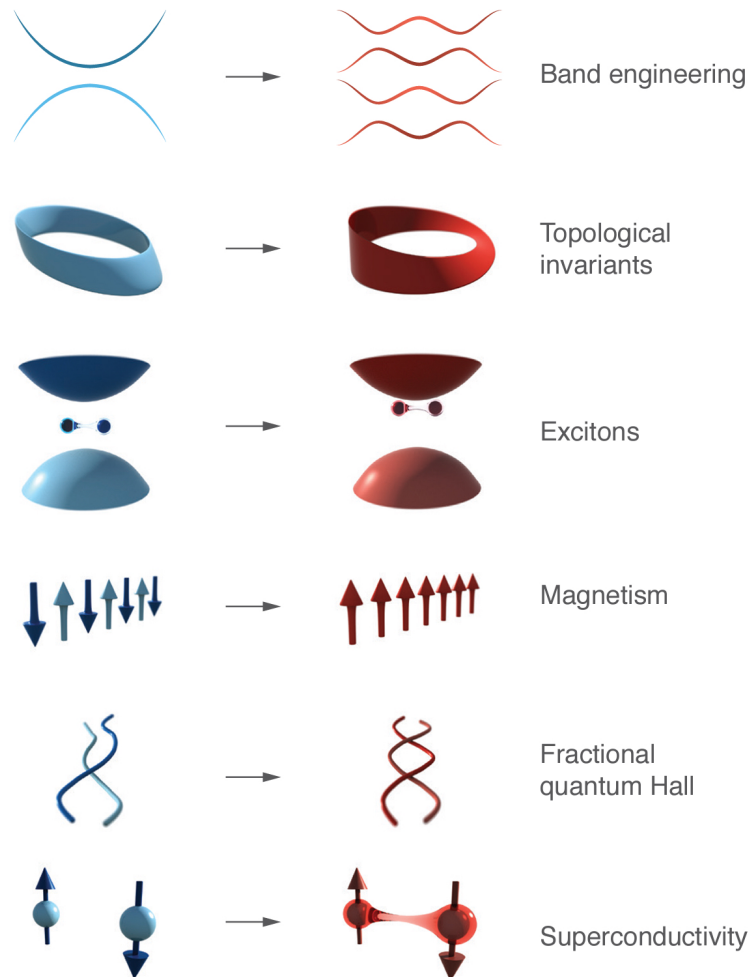
(a) Equilibrium



Optically dressed



(b)

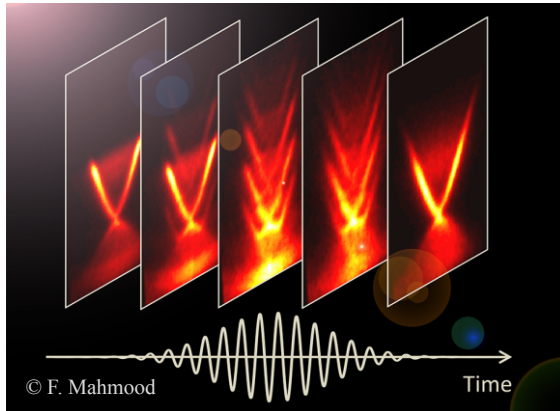


A. de la Torre, D. Kennes, M. Claassen, S. Gerber, J. McIver, MAS, review in prep.

Floquet at work

c.f. Netanel Lindner } talks :i
Isabella Giertz
James McIver

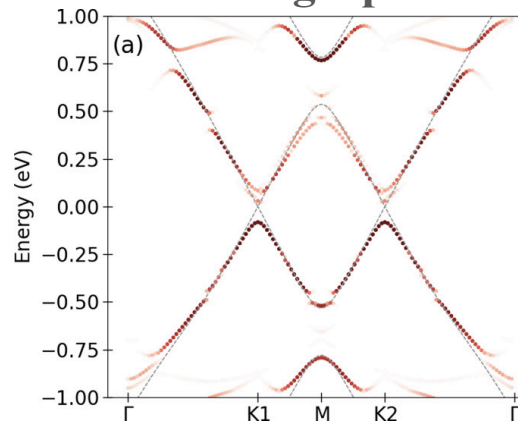
Floquet-Bloch bands in time-resolved ARPES



© F. Mahmood

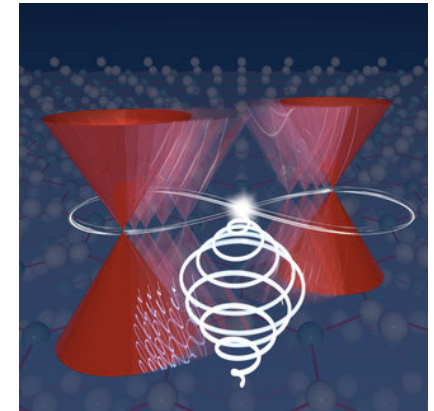
F. Mahmood et al., Nature Physics 12, 306 (2016)

Floquet topology in Moiré graphene

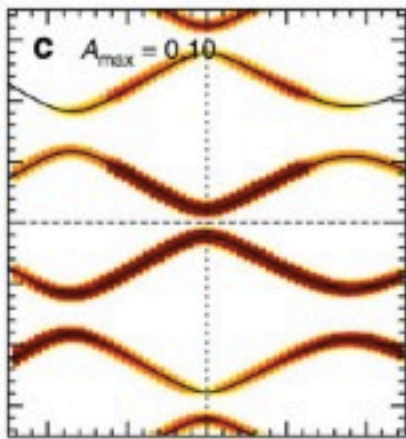


G. E. Topp et al., PRResearch 1, 023031 (2019)

Floquet-Weyl semimetal



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



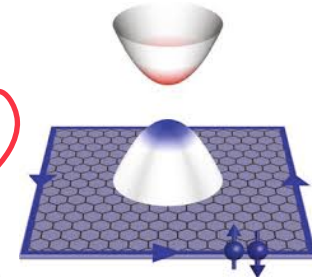
M. A. Sentef et al., Nat. Commun. 6, 7047 (2015)

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

- need for strong lasers
- need for "coherence" (*-> Isabella's talk*)
- detrimental heating effects

possible resolution: **cavities** (next part of talk)

Light-induced anomalous Hall effect



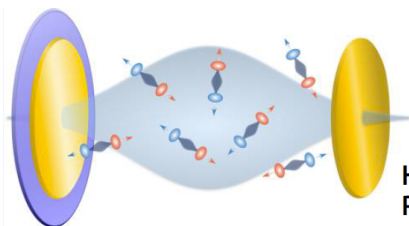
McIver et al., Nat. Phys. 2020

2. Cavity engineering

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



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)

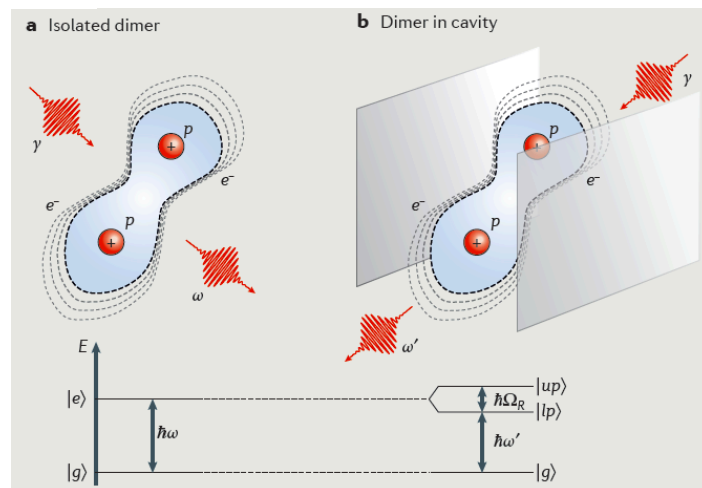
R. F. Ribeiro et al., Chem. Sci. 9, 6325 (2018)

J. Flick et al., Nanophotonics 7, 1479 (2018)

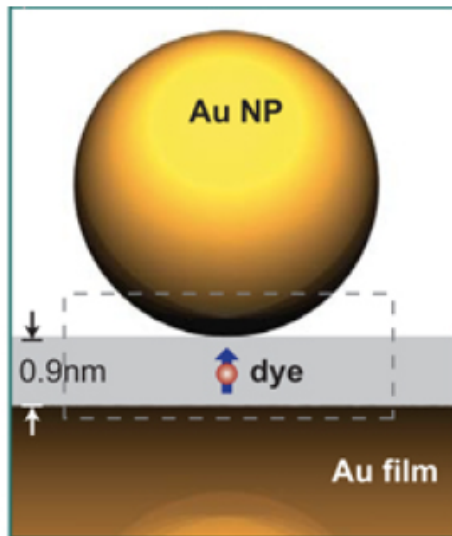
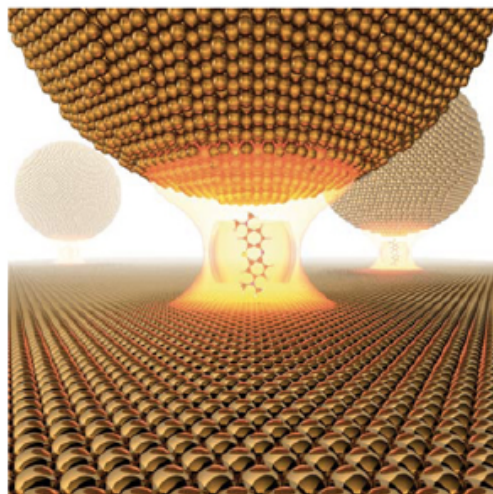
A. F. Kockum et al., Nat. Rev. Phys. 1, 19 (2019)

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**“.



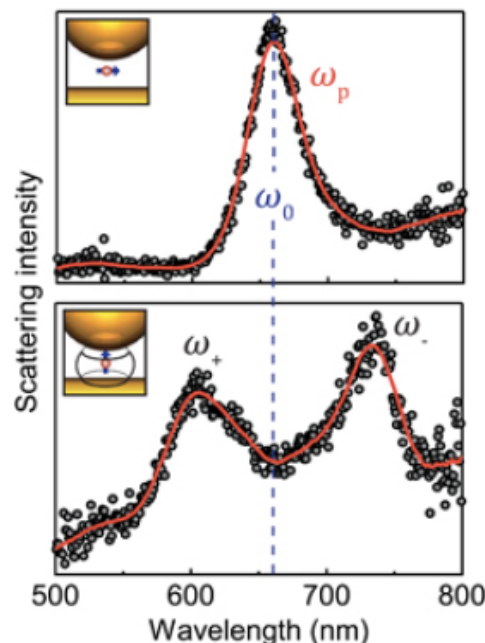
From classical to quantum light



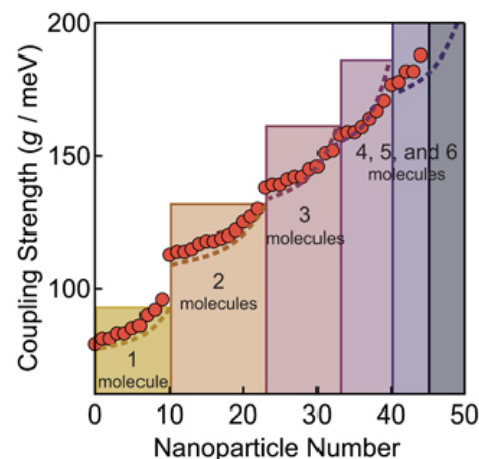
R. Chikkaraddy et al., Nature 535, 127 (2016)

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

cavity materials: many atoms interact with the same modes



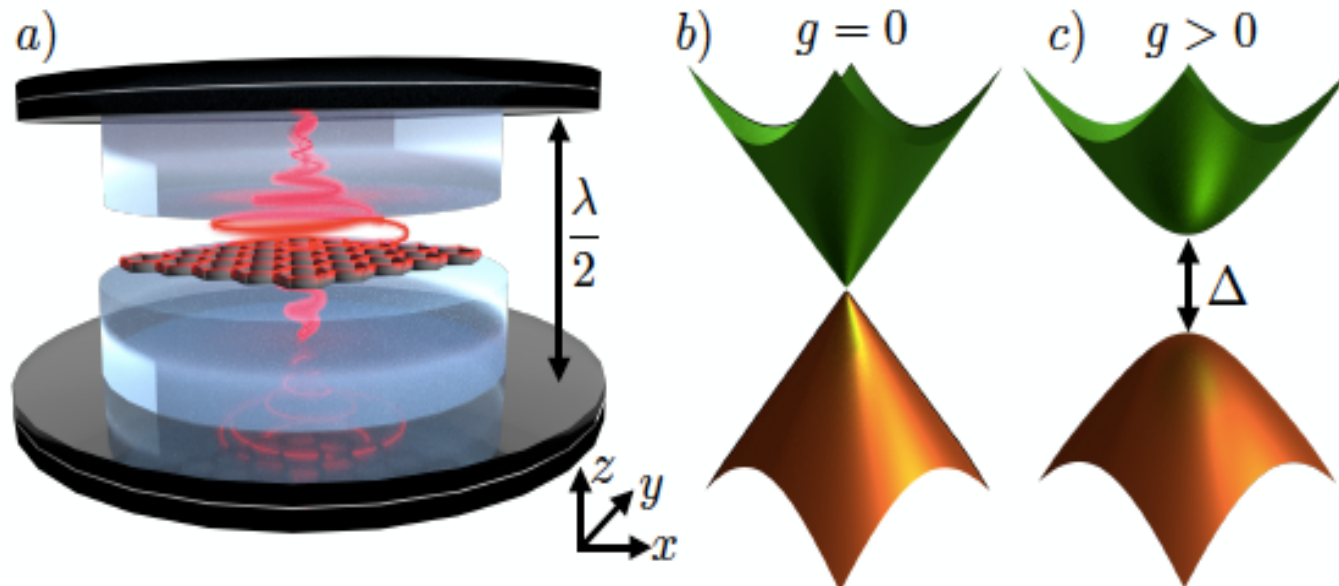
Rabi splitting



Cavity-induced topology

Cavity-induced quantized anomalous Hall effect in graphene

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



Dirac fermion in cavity

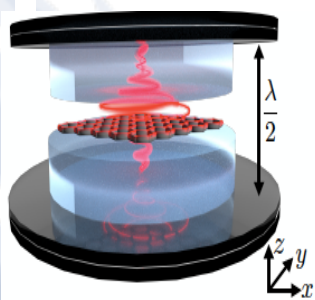
X. Wang et al., PRB 99, 235156 (2019)

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

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

$$\hat{\vec{A}} = A_0 \sum_{\lambda} (\vec{e}_{\lambda} a_{\lambda} + \vec{e}_{\lambda}^* a_{\lambda}^\dagger) \quad A_0 = \sqrt{\hbar / (\epsilon \epsilon_0 V \omega)}$$

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



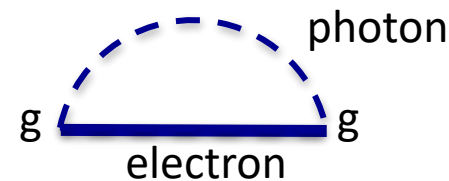
exchange of virtual photons with the cavity vacuum

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, $\gamma(\vec{k} - \hat{A}) \rightarrow \hbar v_F(k_x + ik_y - \sqrt{2}A_0 a^\dagger)$

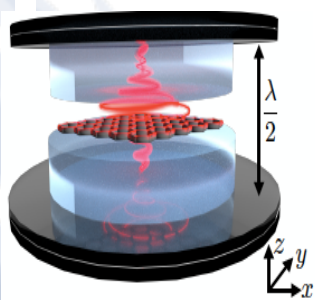
band renormalization due to electron-photon self-energy



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

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

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



Dirac fermion in cavity

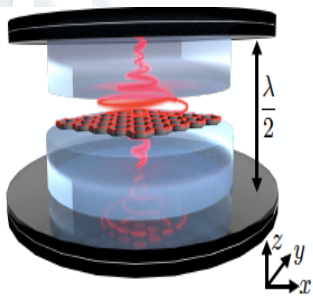
X. Wang et al., PRB 99, 235156 (2019)

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

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

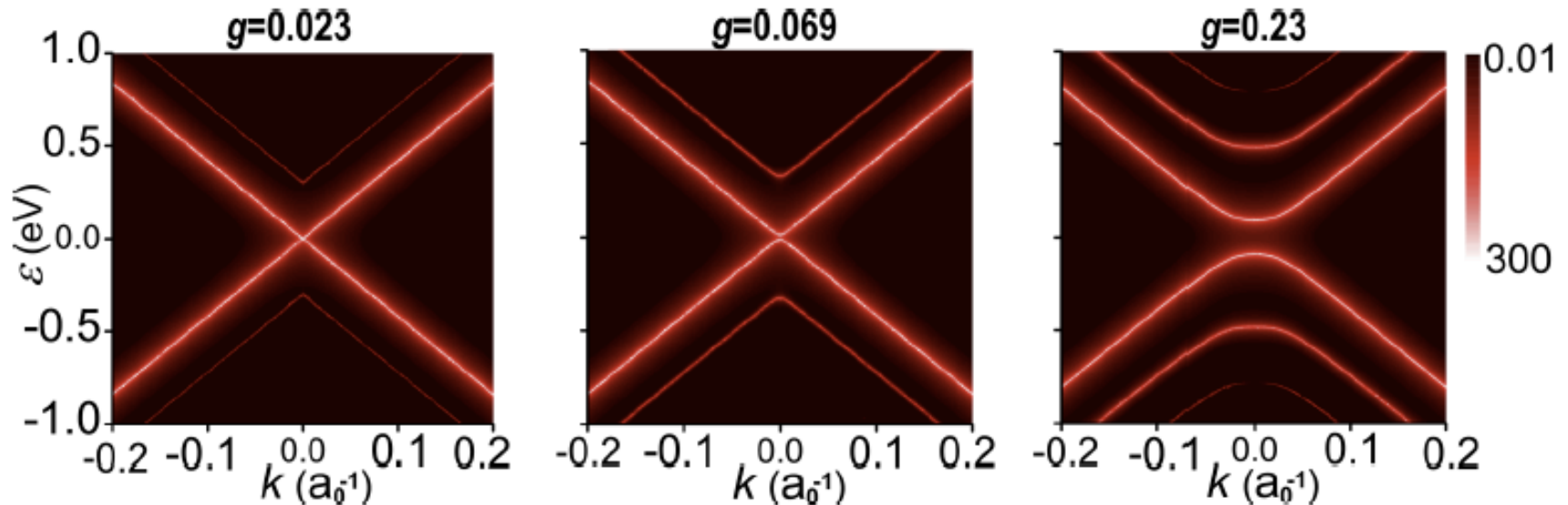
... looks like Floquet result but **different interpretation of A_0** :

- 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

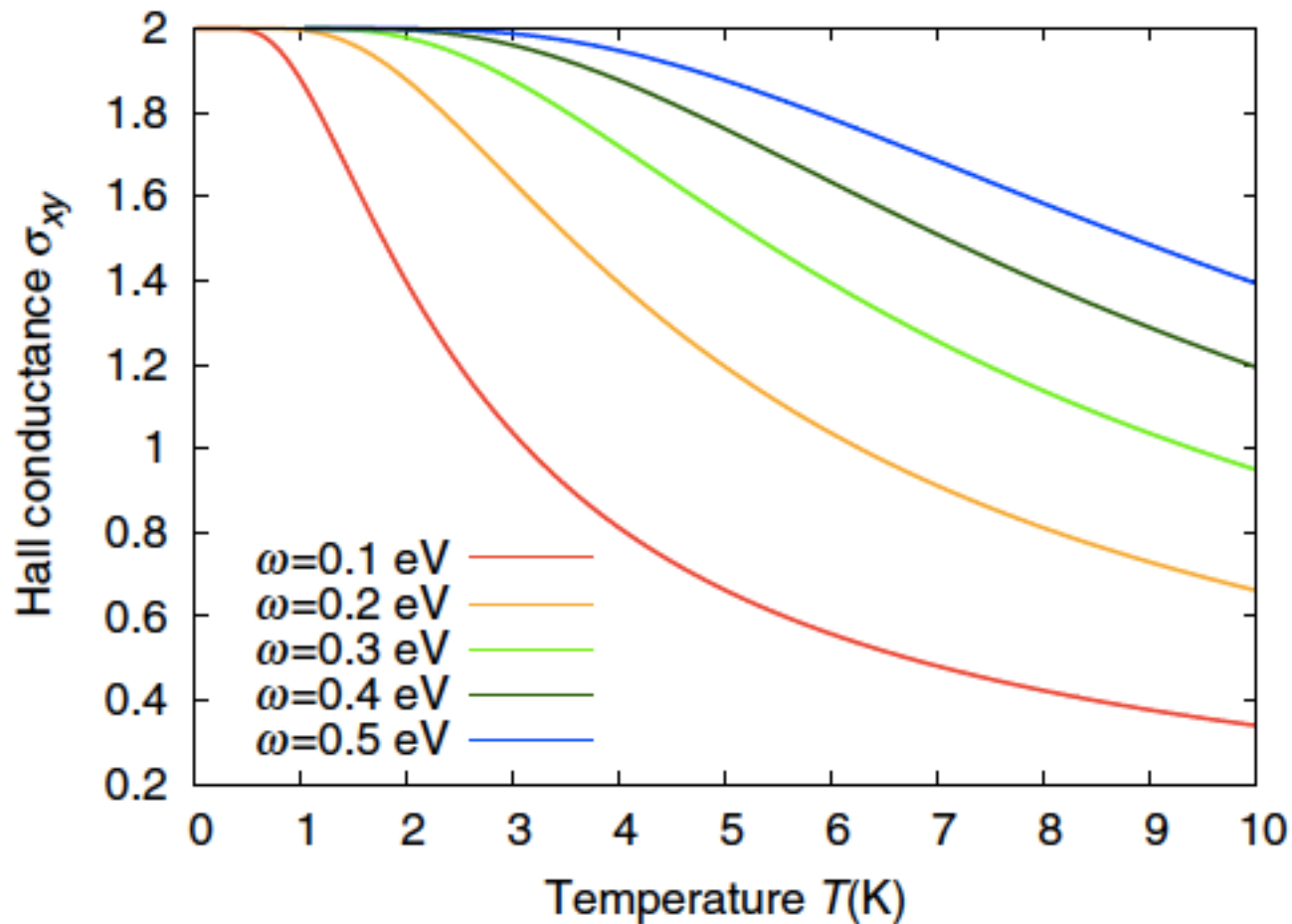
X. Wang et al., PRB 99, 235156 (2019)



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

Dirac fermion in cavity

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

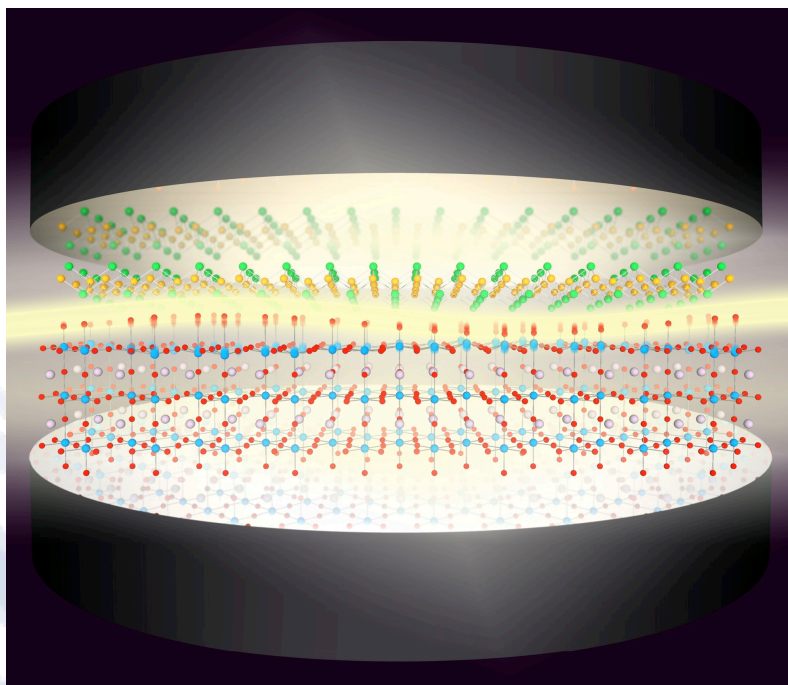


quantized light-induced Hall conductance at low temperatures, controlled by cavity geometry

Cavity superconductivity?

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

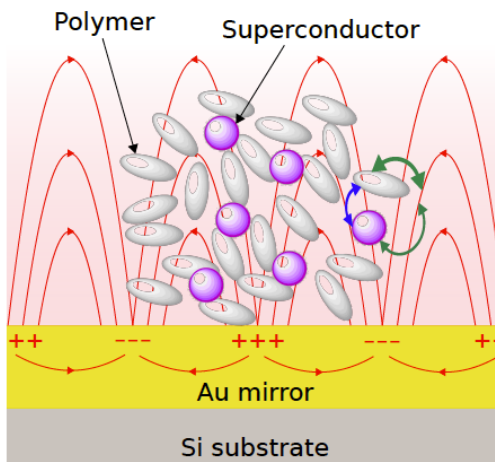
M. A. Sentef, M. Ruggenthaler, A. Rubio, Science Advances 4, eaau6969 (2018)



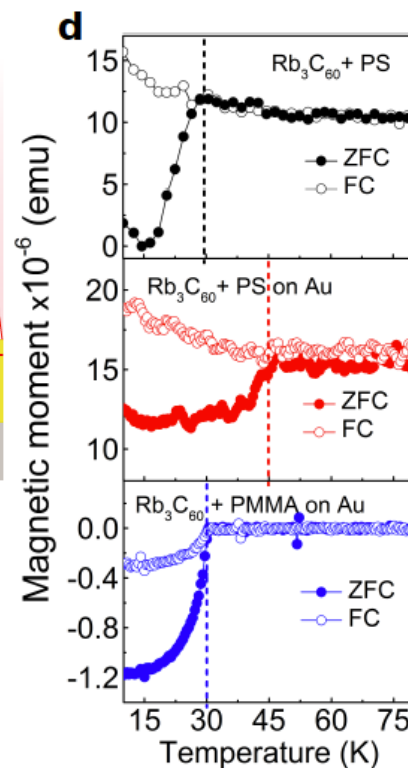
Exploring Superconductivity under Strong Coupling with the Vacuum Electromagnetic Field [arXiv:1911.01459](https://arxiv.org/abs/1911.01459)

A. Thomas¹, E. Devaux¹, K. Nagarajan¹, T. Chervy¹, M. Seidel¹, D. Hagenmüller¹, S. Schütz¹,

J. Schachenmayer¹, C. Genet¹, G. Pupillo^{1*} & T. W. Ebbesen^{1*}



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



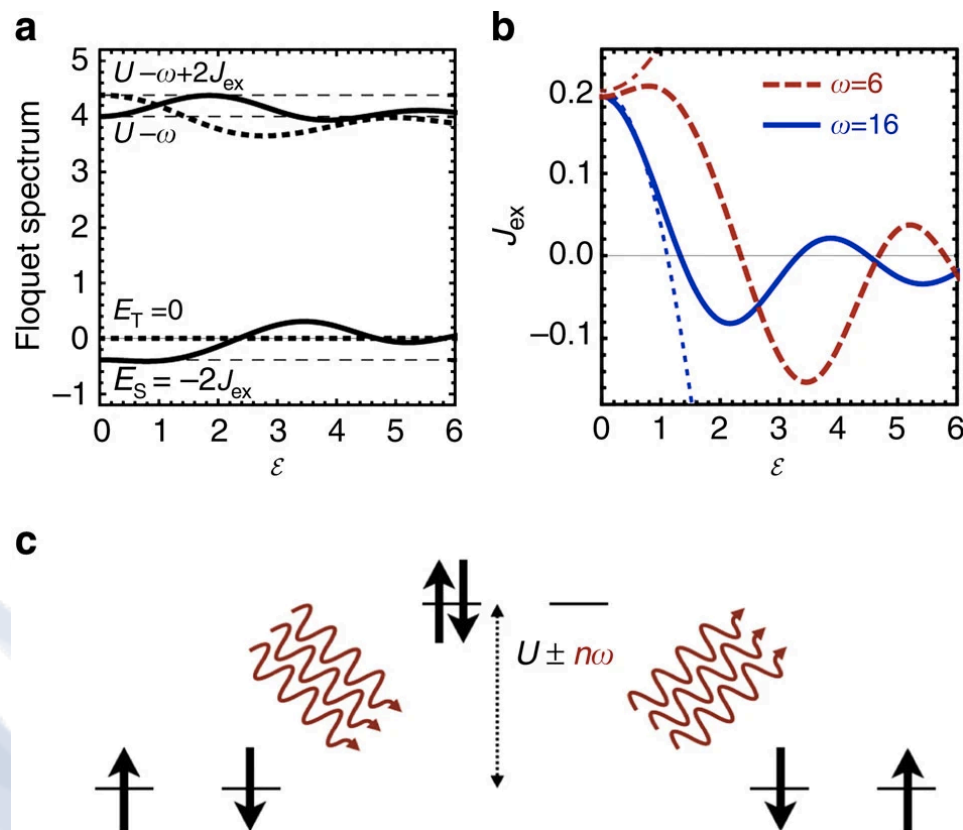
3. Cavity to Floquet crossover

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?

Quantum to classical crossover of Floquet engineering in correlated quantum systemsMichael A. Sentef ^{1,*}, Jiajun Li ², Fabian Künzle ², and Martin Eckstein^{2,†}¹*Max Planck Institute for the Structure and Dynamics of Matter, Luruper Chaussee 149, 22761 Hamburg, Germany*²*Department of Physics, University of Erlangen-Nuremberg, 91058 Erlangen, Germany*

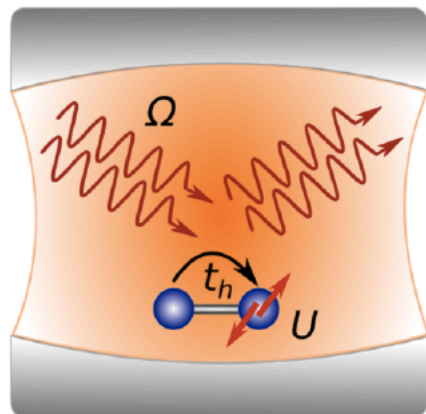
(Received 28 February 2020; accepted 11 June 2020; published 7 July 2020)

Light-matter coupling involving classical and quantum light offers a wide range of possibilities to tune the electronic properties of correlated quantum materials. Two paradigmatic results are the dynamical localization of electrons and the ultrafast control of spin dynamics, which have been discussed within classical Floquet engineering and in the deep quantum regime where vacuum fluctuations modify the properties of materials. Here we discuss how these two extreme limits are interpolated by a cavity which is driven to the excited states. In particular, this is achieved by formulating a Schrieffer-Wolff transformation for the cavity-coupled system, which is mathematically analogous to its Floquet counterpart. Some of the extraordinary results of Floquet engineering, such as the sign reversal of the exchange interaction or electronic tunneling, which are not obtained by coupling to a dark cavity, can already be realized with a single-photon state (no coherent states are needed). The analytic results are verified and extended with numerical simulations on a two-site Hubbard model coupled to a driven cavity mode. Our results generalize the well-established Floquet engineering of correlated electrons to the regime of quantum light. This opens up a pathway of controlling properties of quantum materials with high tunability and low energy dissipation.

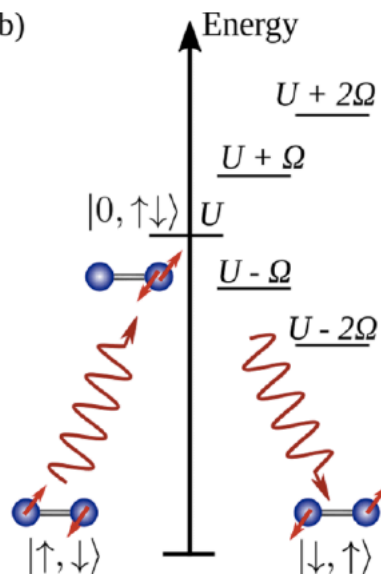
Cavity to Floquet crossover

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

(a)



(b)



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

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

A: effective vector potential

g: light-matter coupling strength

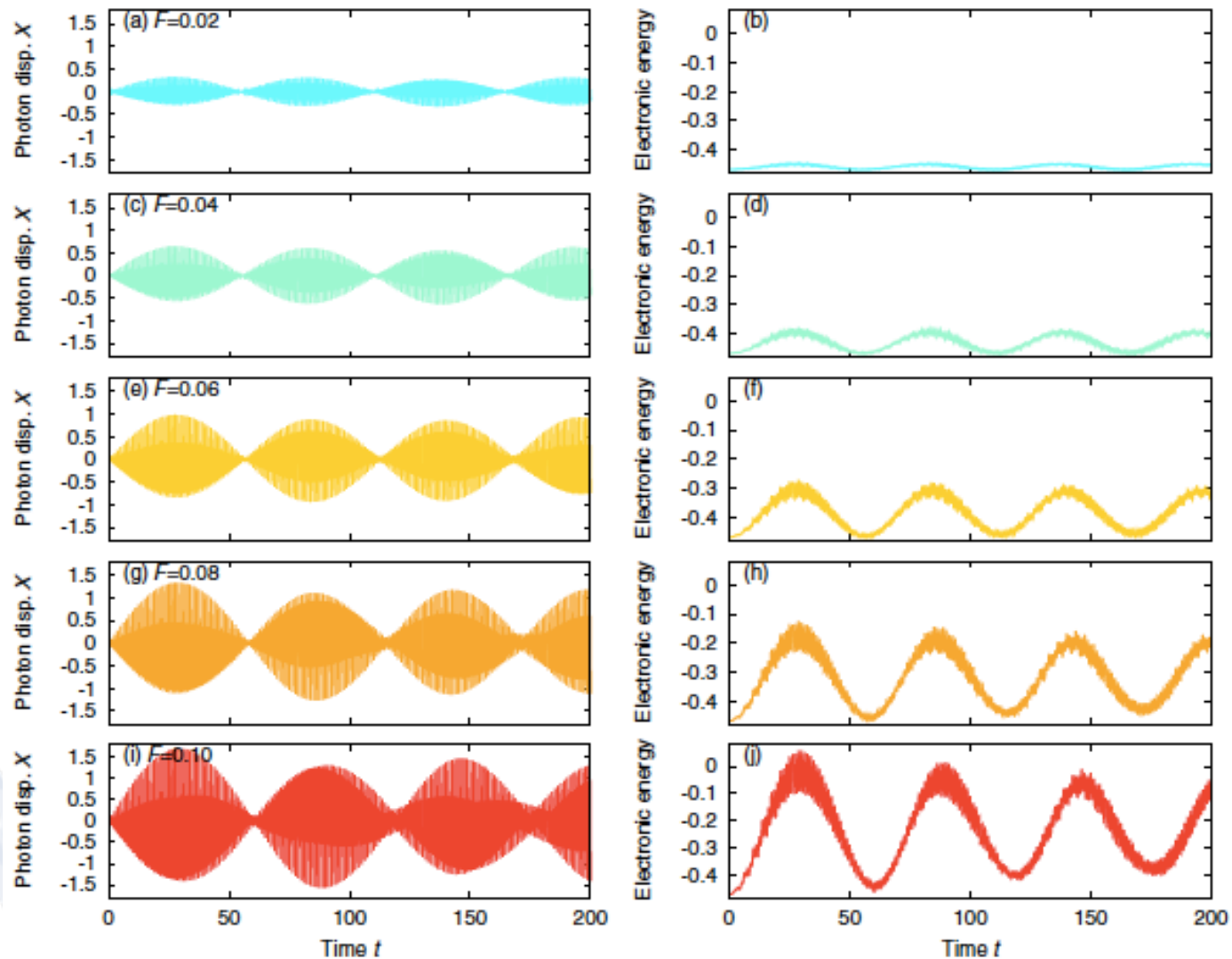
Quantum system -> Floquet system for $n \rightarrow \infty, g\sqrt{n}$ fixed.
(large photon number, weak light-matter coupling strength g)

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

$$\langle \hat{A} \rangle \neq 0$$

Driven cavity

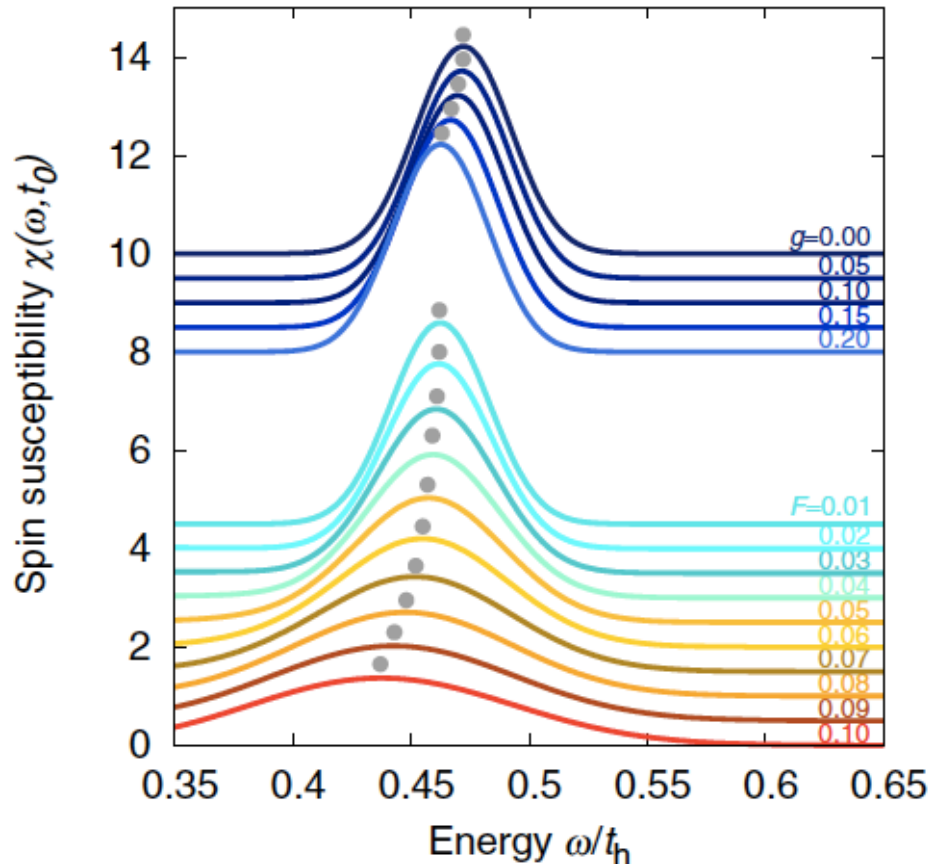
M. A. Sentef, J. Li, F. Künzel, M. Eckstein, PRResearch 2, 033033 (2020)



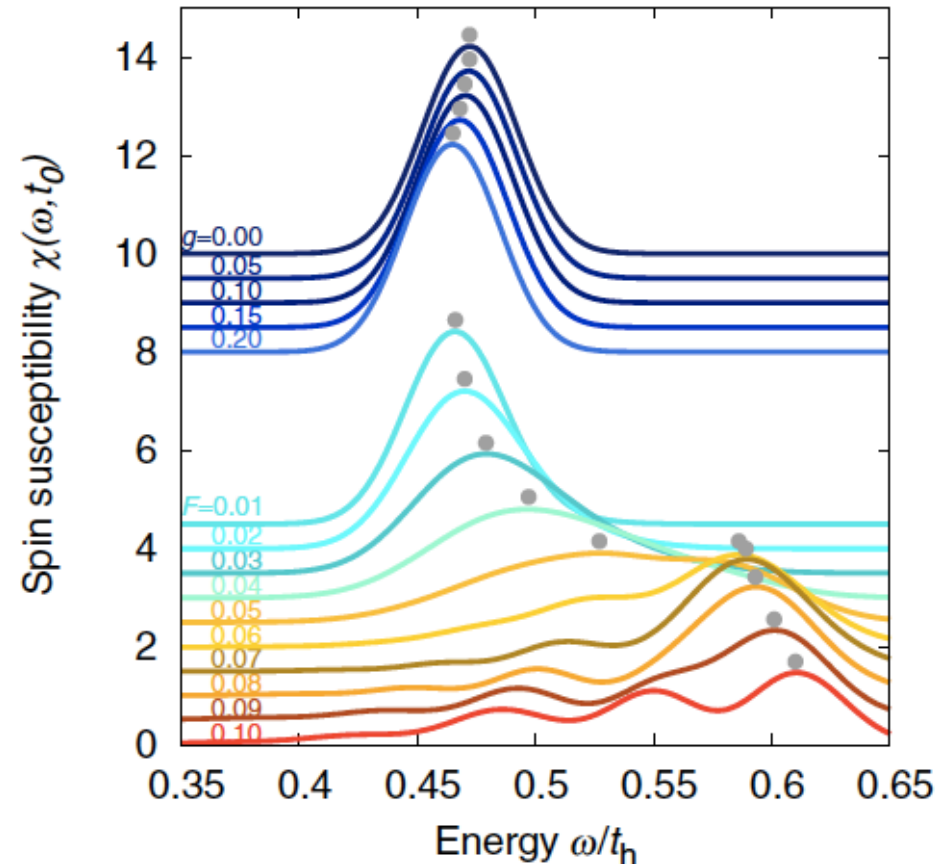
Driven cavity

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

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



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)

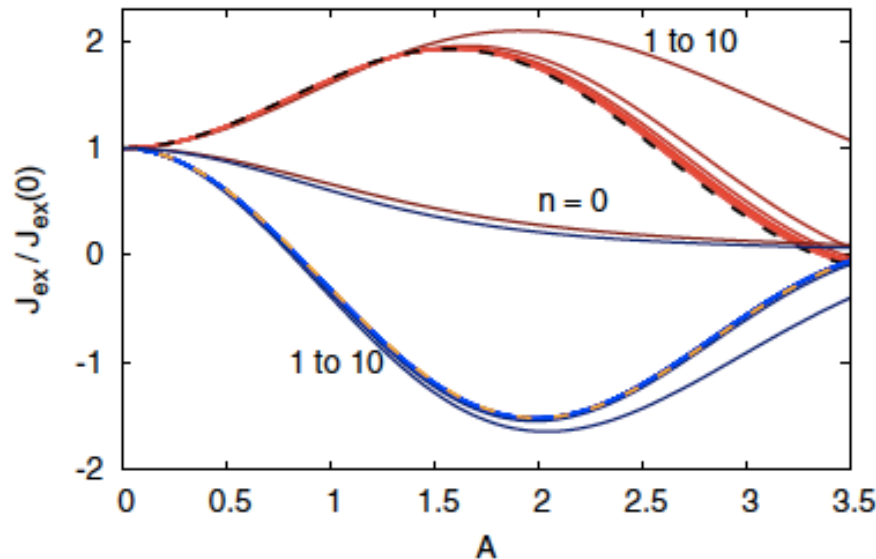


FIG. 4. Exchange interaction $J_{\text{ex}}^{(n)}$ in the n -photon state as a function of A for $n = 1, \dots, 10$. The vertical axis is in units of $J_{\text{ex}}(0)$ at coupling $g = 0$. The curves with colors from dark to light red correspond to $\Omega = 0.8U$ (red-detuned) and those with dark to 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, $J_{\text{ex}}^{(0)}$ 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:

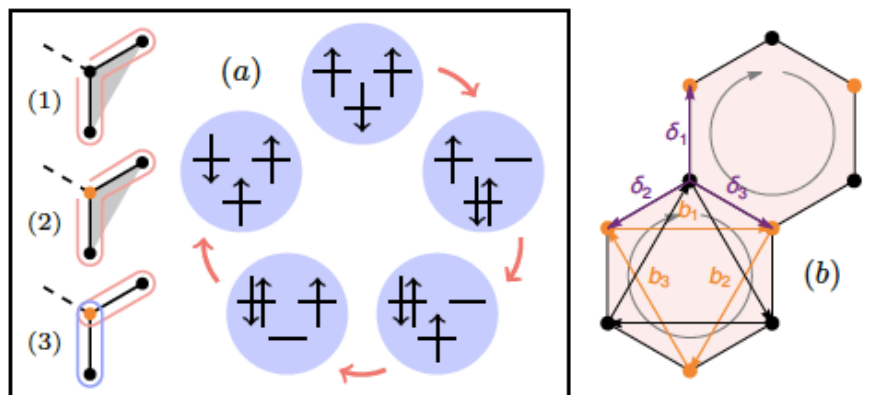
photon number states have zero macroscopic field

-> **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

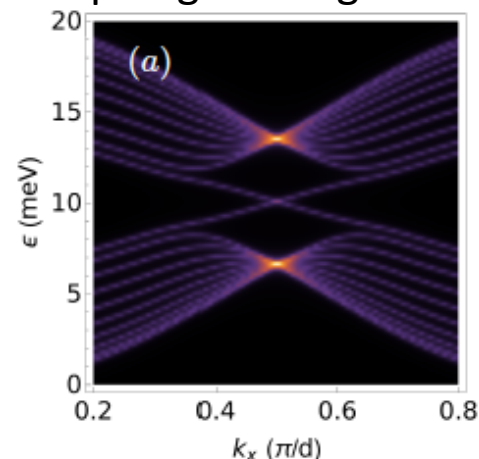
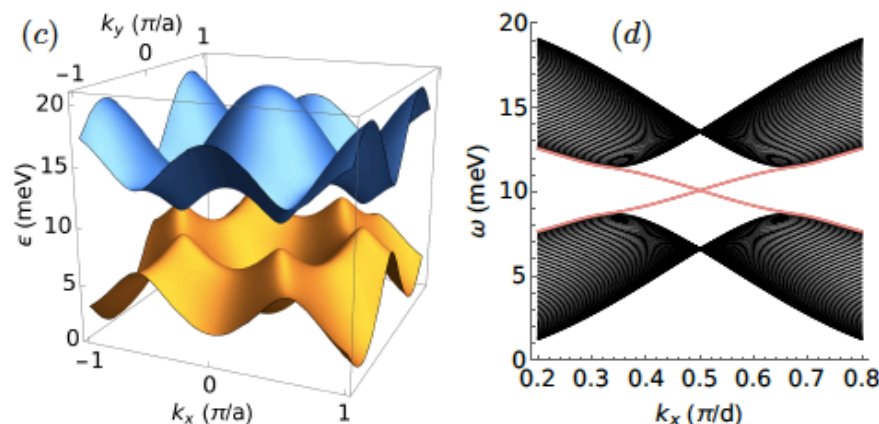
also see: Claassen et al., Nat. Commun. 8, 1192 (2017); Kitamura et al., PRB 96, 014406 (2017); Owerre, Journal of Physics Communications 1, 021002 (2017)



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



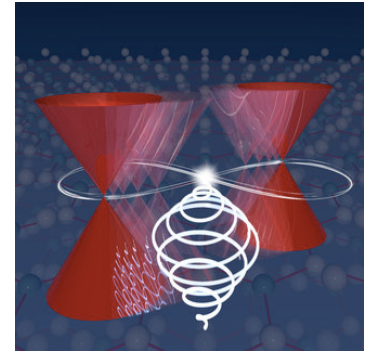
1. Floquet[†] is nice ...

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)



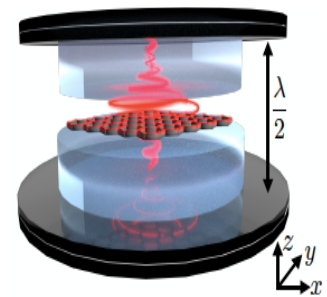
2. ... but cavity materials may be even nicer

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)



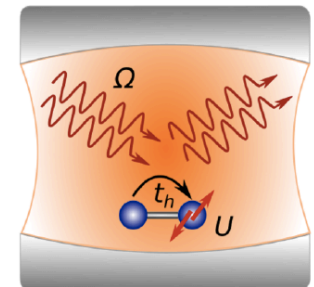
3. ... and there is plenty of room ...

strong light-matter coupling: Floquet effects without coherence

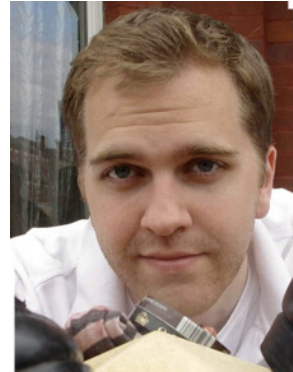
M. A. Sentef, J. Li, F. Künzle, M. Eckstein, PRResearch 2, 033033 (2020)

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

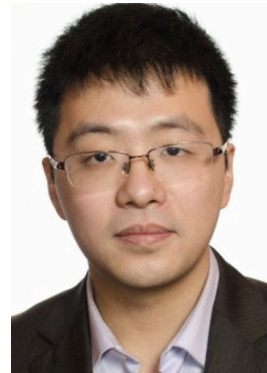
... between the quantum- and classical-
light extremes



Collaborators and Funding



Gabriel Topp Martin Claassen Tom Devereaux Lex Kemper Hannes Hübener Michael Ruggenthaler Ángel Rubio



Gregor Jotzu James McIver Xiao Wang Jiajun Li Martin Eckstein Emil Boström