

# Nonequilibrium Materials Engineering

Michael A. Sentef

*lab.sentef.org*

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

Nijmegen, February 11, 2020



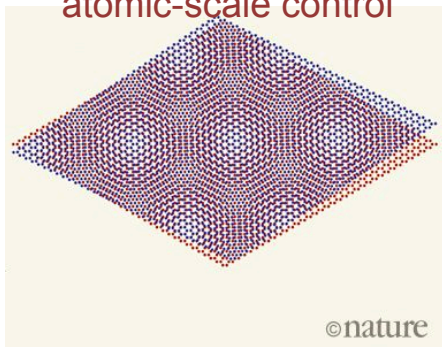
Funded through Deutsche Forschungsgemeinschaft  
Emmy Noether Programme (SE 2558/2-1)

Max Planck Institute for the Structure and Dynamics of Matter



# Engineering materials with light

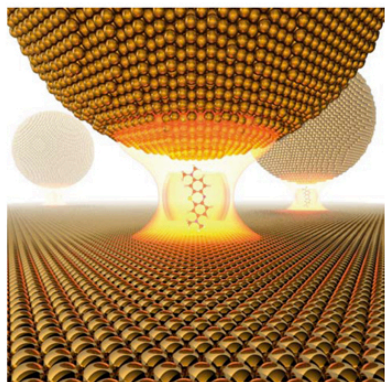
**condensed matter**  
quantum materials  
atomic-scale control



©nature

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

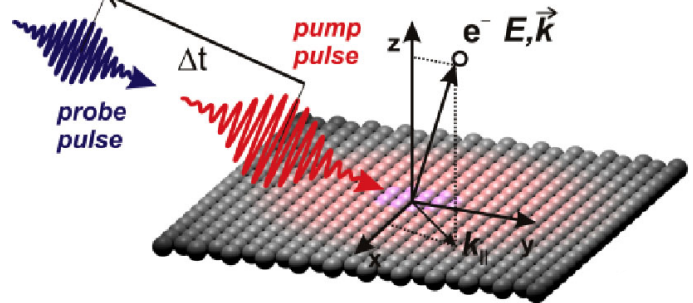
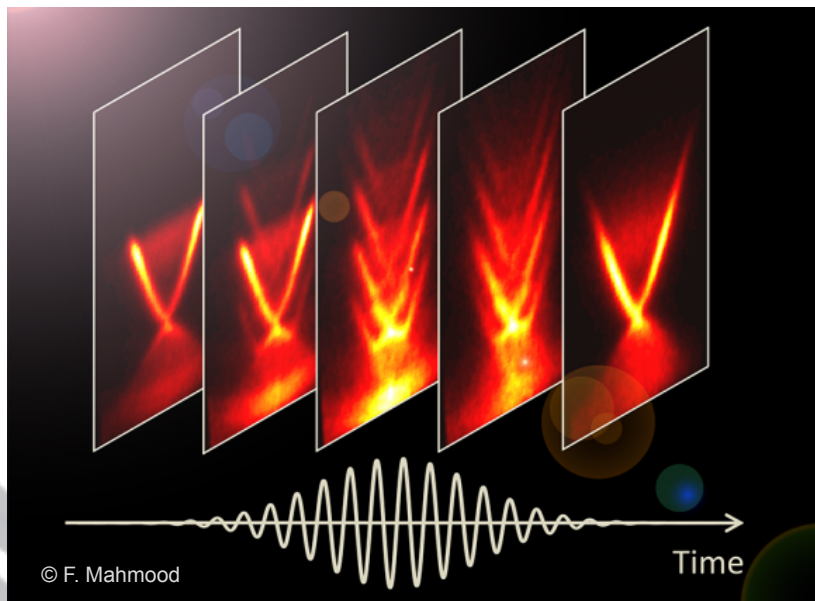


Image courtesy: J. Sobota

**pump-probe: strong classical fields**

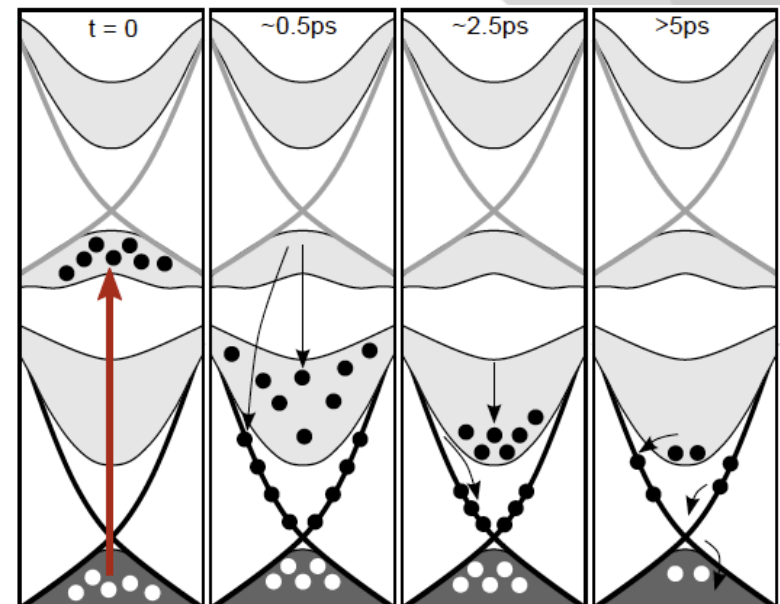
## Hamiltonian engineering

e.g., Floquet-Bloch bands



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

## Distributional engineering



J. Sobota et al., JESRP 195, 249 (2014)

**many ingredients, hard to disentangle**

**this talk:** (I) tailored symmetry breaking, (II) vacuum fluctuations

## How to engineer materials with light?

### Part I: Optical control of chiral superconductors

Short laser pulses allow for switching of Majorana modes

*M. Claassen et al., Nat. Phys. 15, 766 (2019)*

### Part II: From classical to quantized photon fields

Materials engineering in an optical cavity using vacuum fluctuations

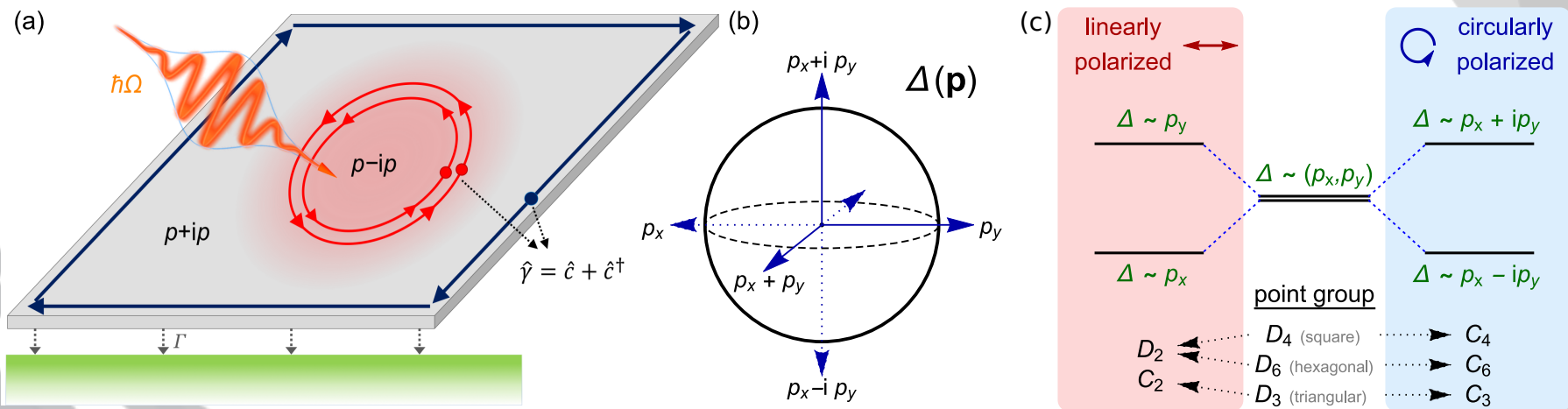
*M. A. Sentef et al., Science Advances 4, eaau6969 (2018)*



# I Optical control of Majoranas

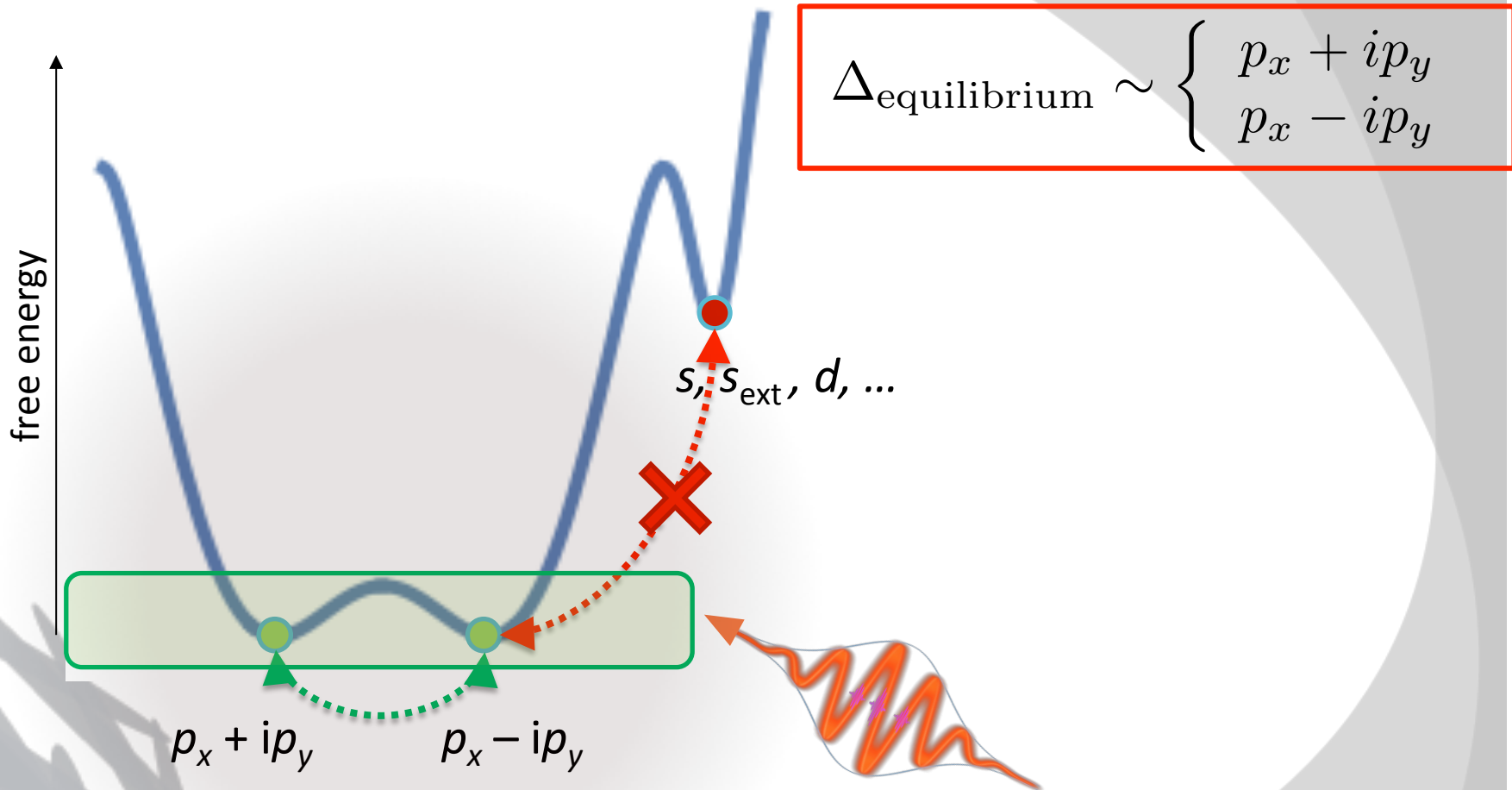
- can one switch the chirality of a 2D topological superconductor?

$\text{Sr}_2\text{RuO}_4$  (?), highly doped graphene, twisted bilayer graphene, ...?



key idea: use two-pulse sequence with linearly and circularly polarized light

# Nonequilibrium pathway to switching



$$\Delta_{\text{equilibrium}} \sim \begin{cases} p_x + ip_y \\ p_x - ip_y \end{cases}$$

$$\Delta_{\text{non-eq}}(t) \sim \cos(\theta) \text{ "}p_x + ip_y\text{"} + \sin(\theta)e^{i\phi} \text{ "}p_x - ip_y\text{"}$$

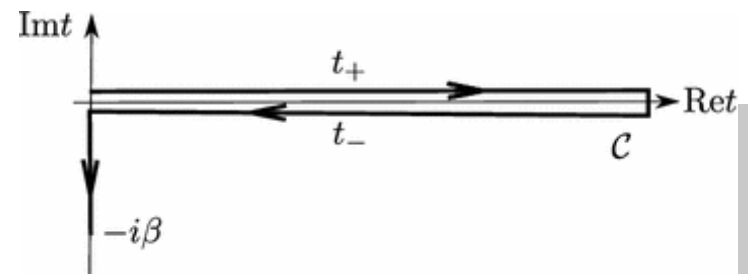
# Model and Method

multiband **Bogoliubov-de-Gennes** Hamiltonians for **doped graphene** (d+id) and **Sr2RuO4** (p+ip)  
coupling to **fermionic reservoir** to dissipate energy  
**laser driving** via Peierls substitution

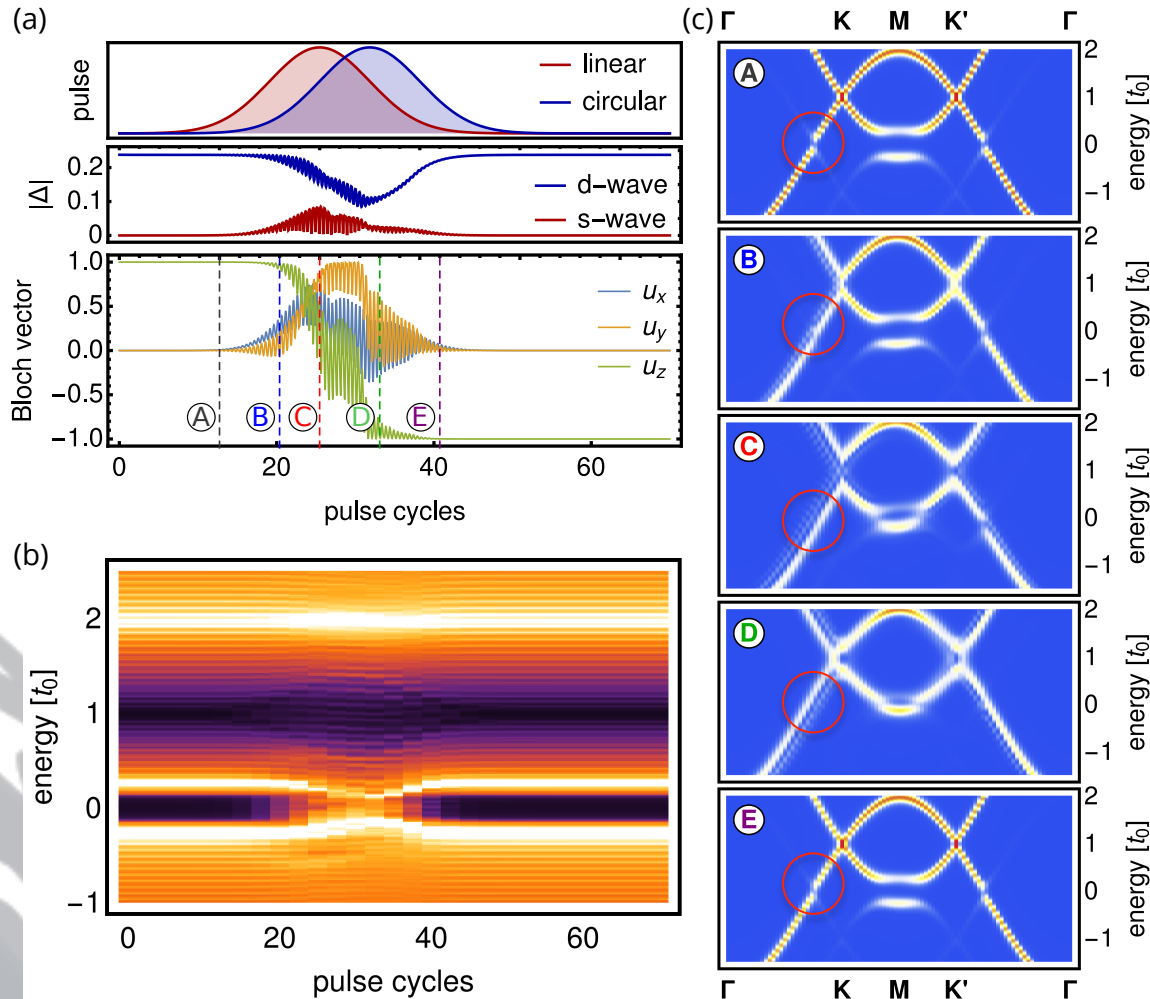
self-consistent Keldysh equations of motion for Nambu Green's functions:

$$i\partial_t \mathcal{G}_{\mathbf{k}}(t, t') = \mathcal{H}_{\mathbf{k}}(t, \Delta_{\mathbf{k}}(t)) \mathcal{G}_{\mathbf{k}}(t, t') + \int d\tau \hat{\Sigma}_{\mathbf{k}}(t, \tau) \mathcal{G}_{\mathbf{k}}(\tau, t')$$

$$\Delta_{\mathbf{k}}(t) = \frac{1}{L} \sum_j v^{(j)} \hat{\eta}_{\mathbf{k}}^{(j)} \sum_{\substack{\mathbf{k}' \\ \alpha\beta}} \hat{\eta}_{\mathbf{k}'\alpha\beta}^{(j)} \left\langle \hat{c}_{-\mathbf{k}',\beta\downarrow} \hat{c}_{\mathbf{k}',\alpha\uparrow} \right\rangle$$



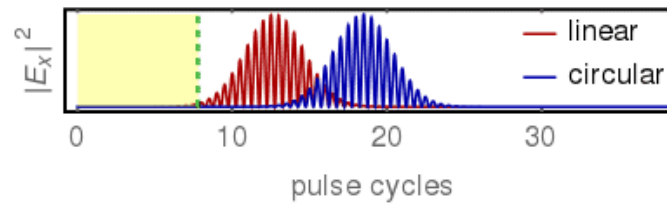
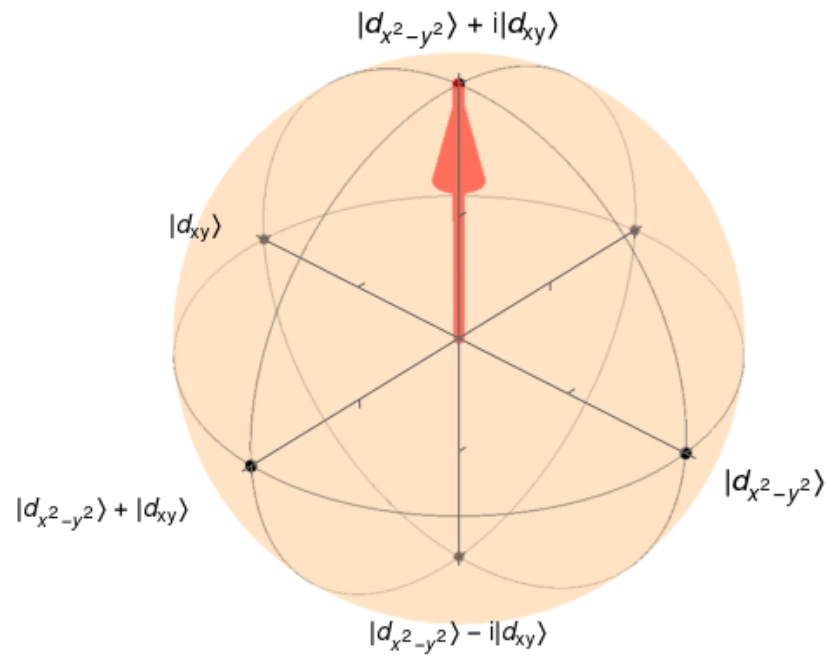
# Optical control of Majoranas



two-pulse sequence  
reverses d+id state  
in graphene

time-resolved  
spectroscopy tracks  
chirality reversal

# Bloch vector rotation

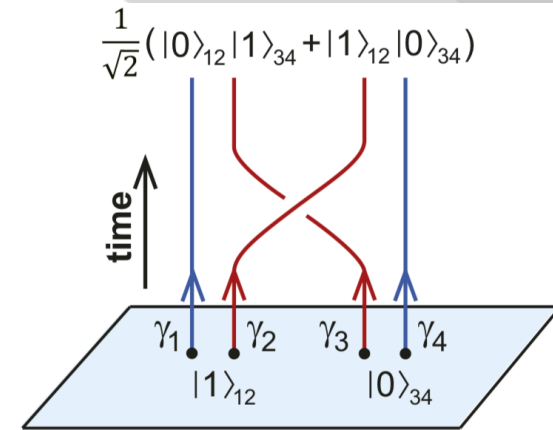




# A „programmable“ topological quantum computer?

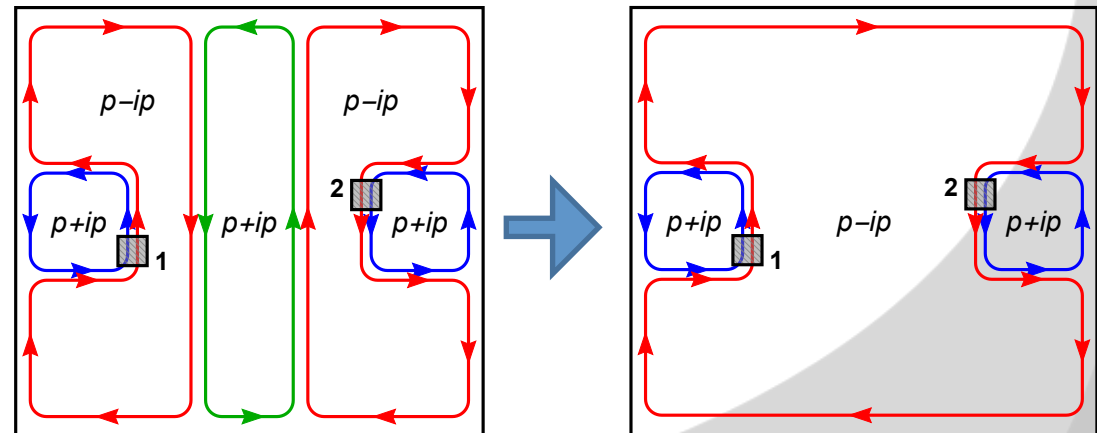
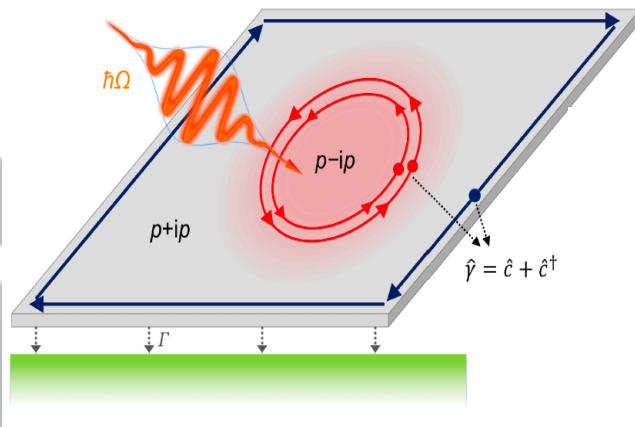
## non-Abelian statistics of Majorana fermions:

- half-quantum vortices of chiral superconductors host single Majorana fermions
  - Two Majoranas represent one electron:  $\frac{1}{2} + \frac{1}{2} = 1$
- Braiding between Majoranas is a non-Abelian operation in electron (charge) basis!



Ivanov, PRL 86, 268 (2001)  
B. Lian et al., PNAS 115, 10938 (2018)

simplest operation: a **switchable Hadamard gate**



# Summary I

- All-optical **control of chiral Majorana modes**
- towards arbitrarily programmable quantum computer?

„program the gate optically, read it out electrically“

*M. Claassen et al.,  
Nat. Phys. 15, 766 (2019)*



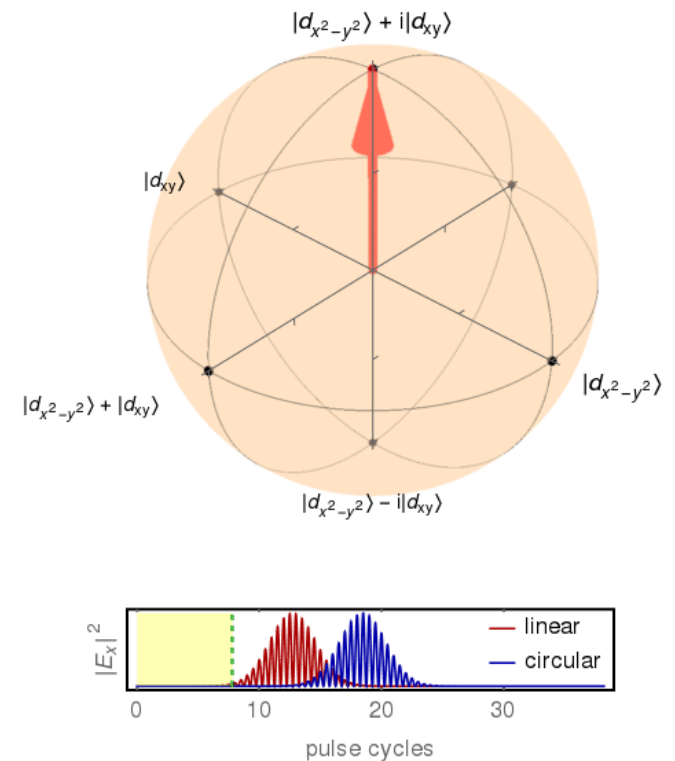
M. Claassen



D. Kennes



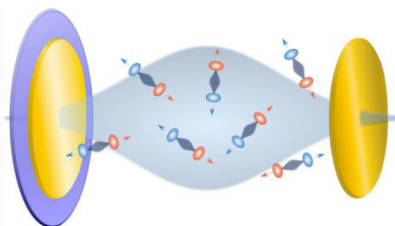
M. Zingl



## 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)*

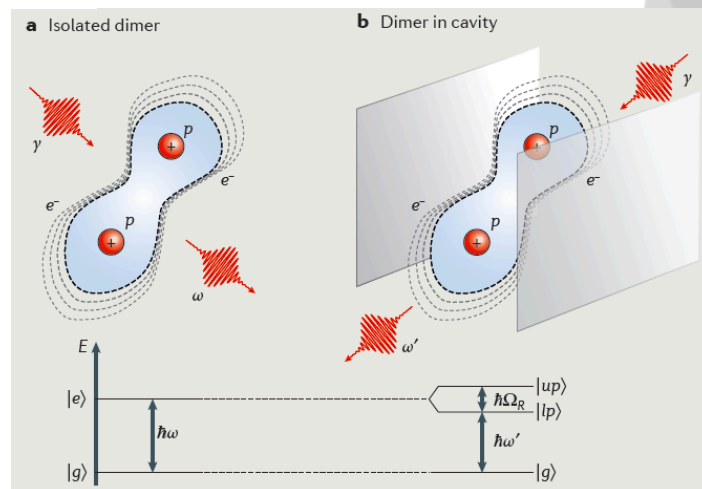
*R. F. Ribeiro et al., Chem. Sci. 9, 6325 (2018)*

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

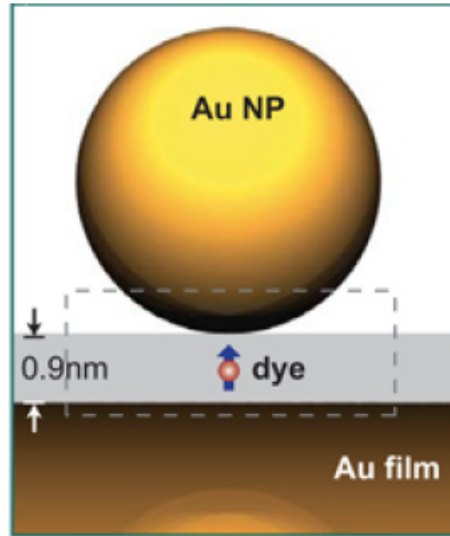
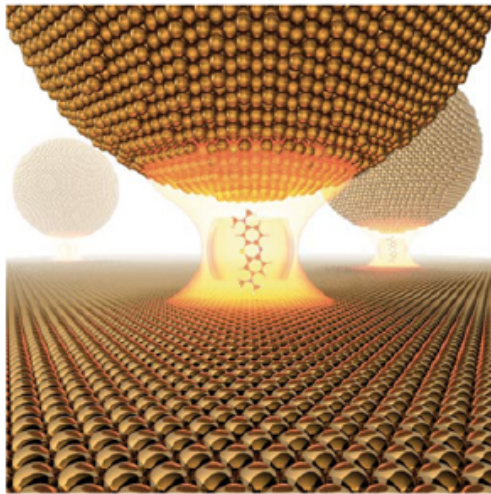
*A. F. Kockum et al., Nat. Rev. Phys. 1, 19 (2019)*

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**“.



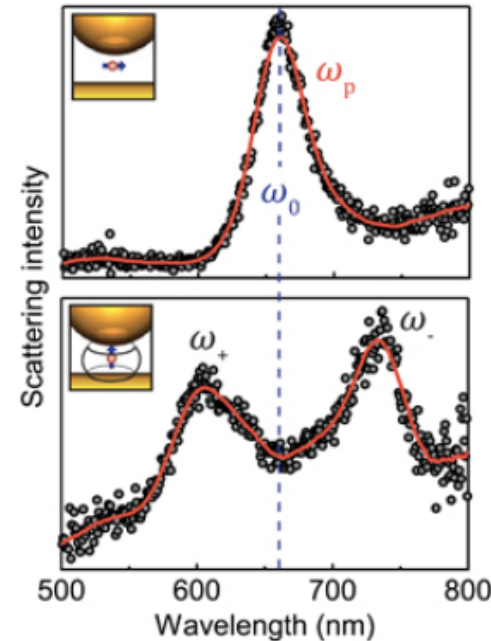
# From classical to quantum light



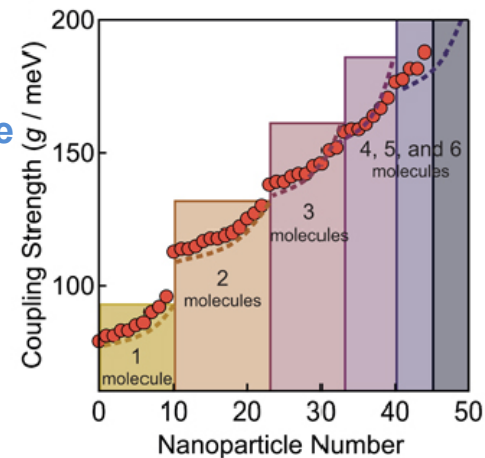
R. Chikkaraddy et al., Nature 535, 127 (2016)

**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





*BCS superconductors: phonon-mediated superconductivity*

*Ginzburg, Phys. Lett. 13, 101 (1964): exciton-mediated superconductivity?*

*Ruvalds, Phys. Rev. B 35, 8869(R) (1987): plasmon-mediated superconductivity?*

PRL 104, 106402 (2010)

PHYSICAL REVIEW LETTERS

week ending  
12 MARCH 2010

Exciton-Polariton Mediated Superconductivity

Fabrice P. Laussy,<sup>1</sup> Alexey V. Kavokin,<sup>1,2</sup> and Ivan A. Shelykh<sup>3,4</sup>

**Cavity-assisted mesoscopic transport of fermions:  
Coherent and dissipative dynamics.**

*Hagenmüller et al., 1801.09876*

**Cavity-mediated electron-photon superconductivity**

Frank Schlawin<sup>1</sup>, Andrea Cavalleri<sup>1,2</sup> and Dieter Jaksch<sup>1</sup>

*1804.07142*

**Cavity Quantum Eliashberg Enhancement of Superconductivity**

Jonathan B. Curtis,<sup>1,2,\*</sup> Zachary M. Raines,<sup>1,2</sup> Andrew A. Allocca,<sup>1,2</sup> Mohammad Hafezi,<sup>1</sup> and Victor M. Galitski<sup>1,2</sup>

*1805.01482*

**Manipulating quantum materials with quantum light**

Martin Kiffner<sup>1,2</sup>, Jonathan Coulthard<sup>2</sup>, Frank Schlawin<sup>2</sup>, Arzhang Ardavan<sup>2</sup>, and Dieter Jaksch<sup>2,1</sup>

*1806.06752*

**Cavity superconductor-polaritons** *1807.06601*

Andrew A. Allocca,<sup>\*</sup> Zachary M. Raines, Jonathan B. Curtis, and Victor M. Galitski

PHYSICAL REVIEW B 93, 054510 (2016)

**Superconductivity and other collective phenomena in a hybrid Bose-Fermi mixture formed by a polariton condensate and an electron system in two dimensions**

Ovidiu Cotelet,<sup>1,\*</sup> Sina Zeytinoglu,<sup>1,2</sup> Manfred Sigrist,<sup>2</sup> Eugene Demler,<sup>3</sup> and Ataç Imamoglu<sup>1</sup>

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

M. A. Sentef,<sup>1,\*</sup> M. Ruggenthaler,<sup>1</sup> and A. Rubio<sup>1,2,3</sup>

*1802.09437*

**Superradiant Quantum Materials**

Giacomo Mazza<sup>1,2,\*</sup> and Antoine Georges<sup>2,3,1,4</sup>

*1804.08534*

Ab-initio Exciton-polaritons:

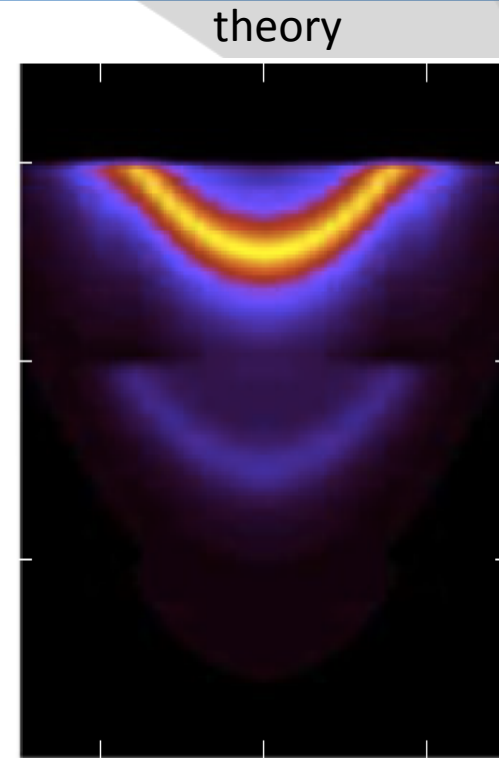
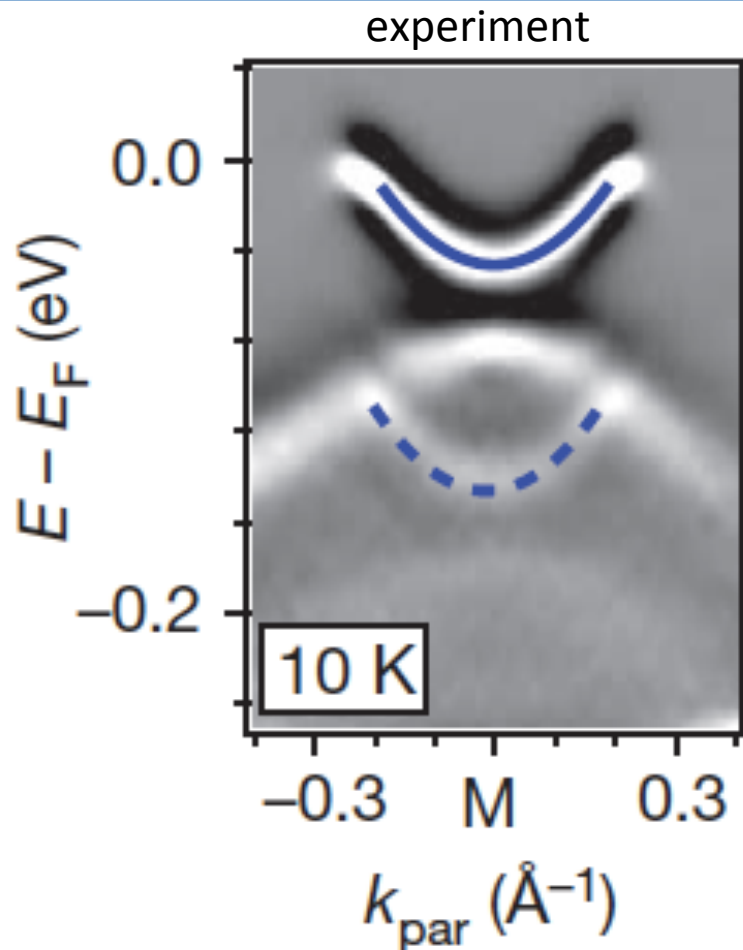
Cavity control of Dark Excitons in two dimensional Materials

Simone Latini,<sup>1,\*</sup> Enrico Ronca,<sup>1,†</sup> Umberto De Giovannini,<sup>1,2,‡</sup> Hannes Hübenner,<sup>1,§</sup> and Angel Rubio<sup>1,3,¶</sup>

*1810.02672*



# monolayer FeSe/STO: ARPES



replica bands: forward (small- $q$ )  
electron-phonon scattering

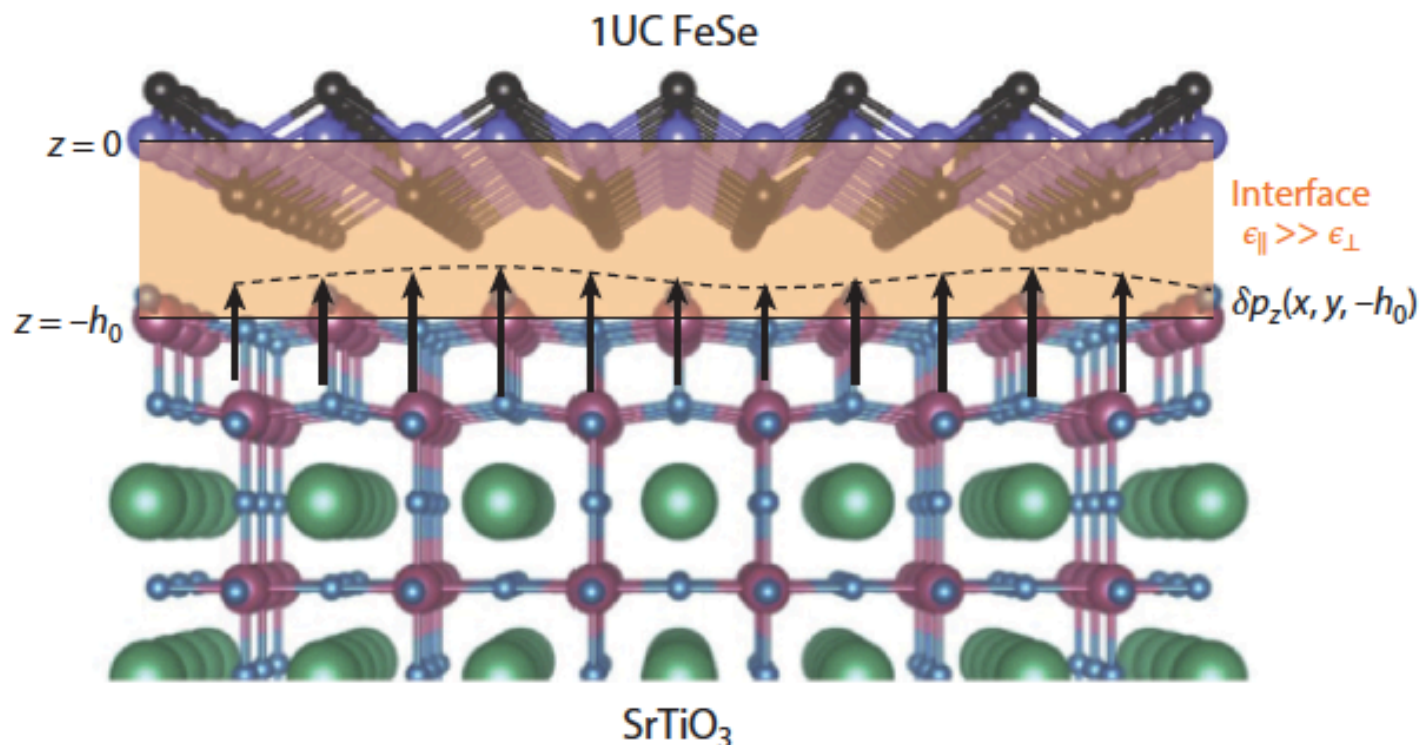
*Lee et al., Nature 515, 245 (2014)*

*Rademaker et al., New J. Phys. 18, 022001 (2016)*

# monolayer FeSe/STO: interfacial phonon

bare el-phonon vertex  $g(\vec{q}) = g_0 \exp(-|\vec{q}|/q_0)$  *Lee et al., Nature 515, 245 (2014)*

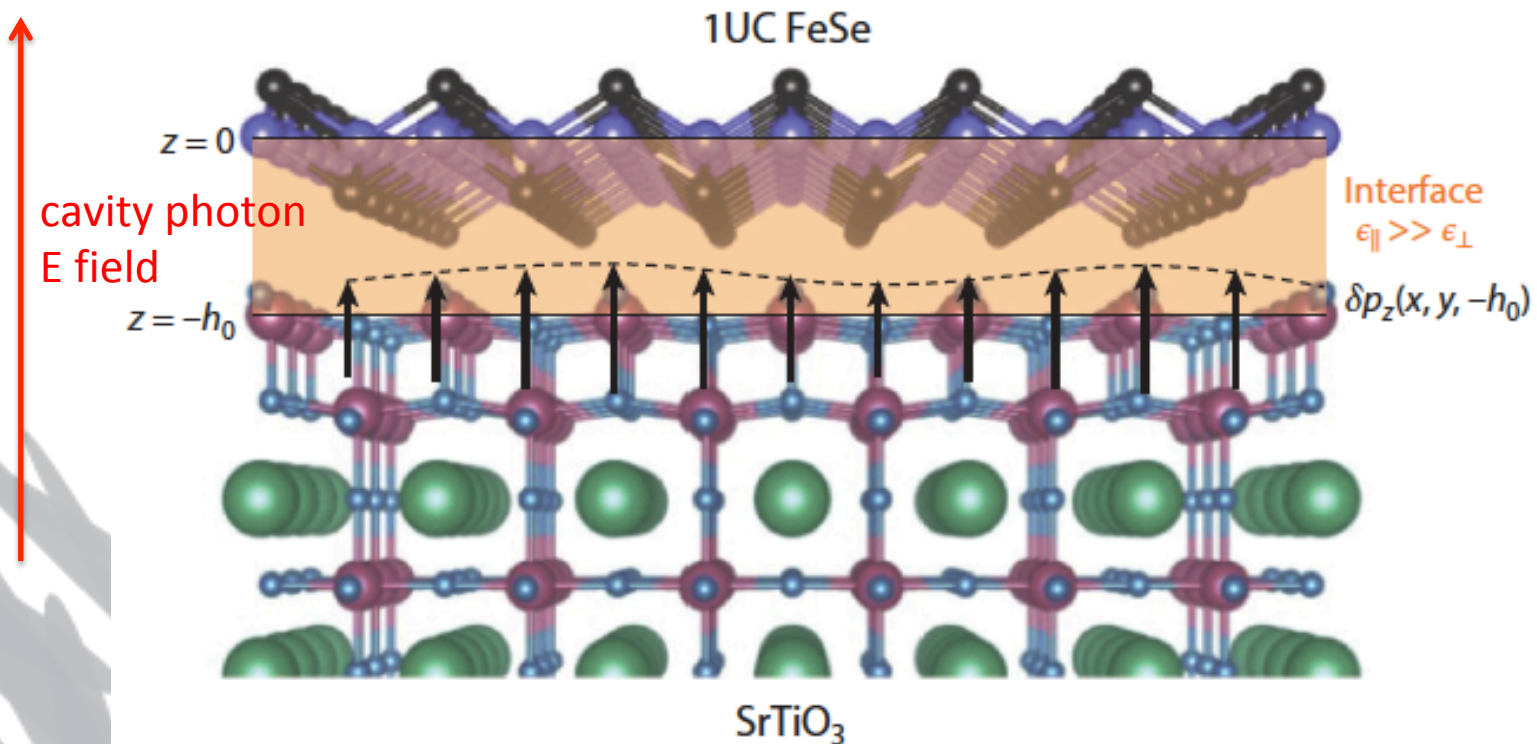
$$q_0^{-1} = h_0 \sqrt{\epsilon_{\parallel}/\epsilon_{\perp}} \quad \epsilon_{\parallel}/\epsilon_{\perp} \approx 100$$



*Huang and Hoffman, Annu. Rev. CMP 8, 311 (2017)*

# Cavity engineering

- idea: use **phonon polaritons** to modify electron-phonon coupling



*Huang and Hoffman, Annu. Rev. CMP 8, 311 (2017)*

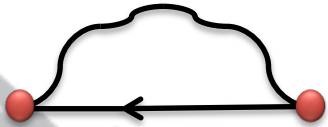
$$H = \sum_{\vec{k}, \sigma} \epsilon_{\vec{k}} c_{\vec{k}, \sigma}^\dagger c_{\vec{k}, \sigma} + \frac{1}{\sqrt{N}} \sum_{\vec{k}, \vec{q}, \sigma, \lambda = \pm} c_{\vec{k} + \vec{q}, \sigma}^\dagger c_{\vec{k}, \sigma} (g_\lambda^*(\vec{q}) \alpha_{-\vec{q}, \lambda}^\dagger + g_\lambda(\vec{q}) \alpha_{\vec{q}, \lambda}) + \sum_{\vec{q}, \lambda = \pm} \omega_\lambda(\vec{q}) \alpha_{\vec{q}, \lambda}^\dagger \alpha_{\vec{q}, \lambda}$$

electrons                      el-polariton coupling                      polaritons

bare el-phonon vertex     $g(\vec{q}) = g_0 \exp(-|\vec{q}|/q_0)$      $q_0^{-1} = h_0 \sqrt{\epsilon_{\parallel} / \epsilon_{\perp}}$

G-self-consistent Migdal-Eliashberg diagram

$$\hat{\Sigma}(\vec{k}, i\omega_n) = \frac{-1}{N\beta} \sum_{\vec{q}, m, \lambda = \pm} |g_\lambda(\vec{q})|^2 D_\lambda^{(0)}(\vec{q}, i\omega_n - i\omega_m) \hat{\tau}_3 \hat{G}(\vec{k} + \vec{q}, i\omega_m) \hat{\tau}_3$$

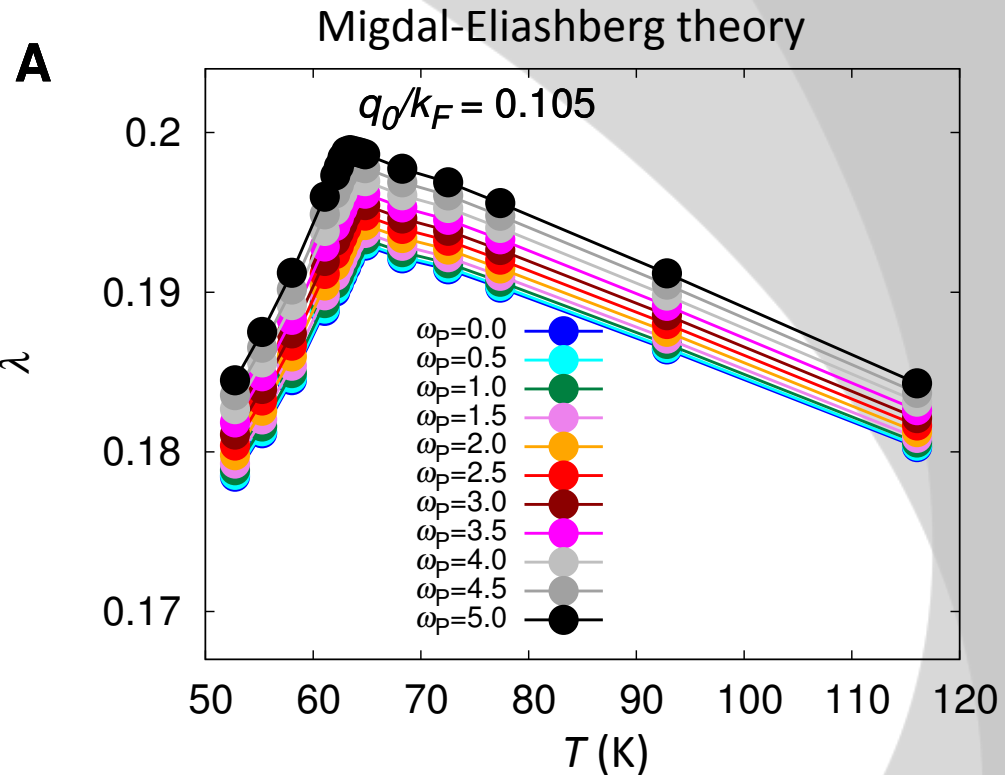
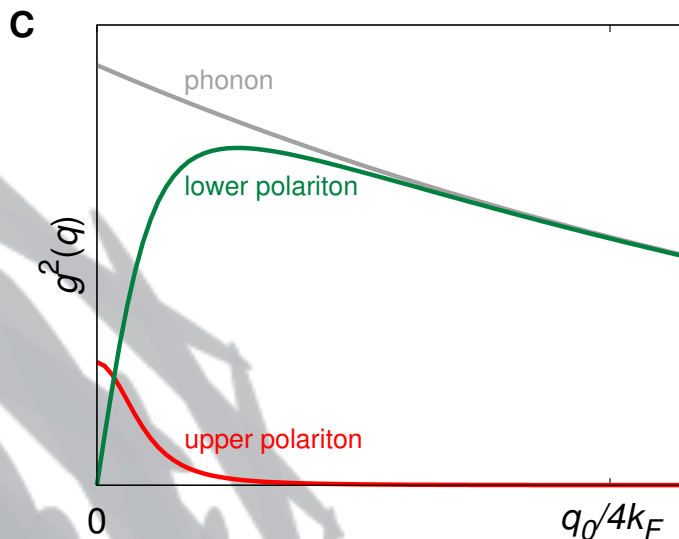
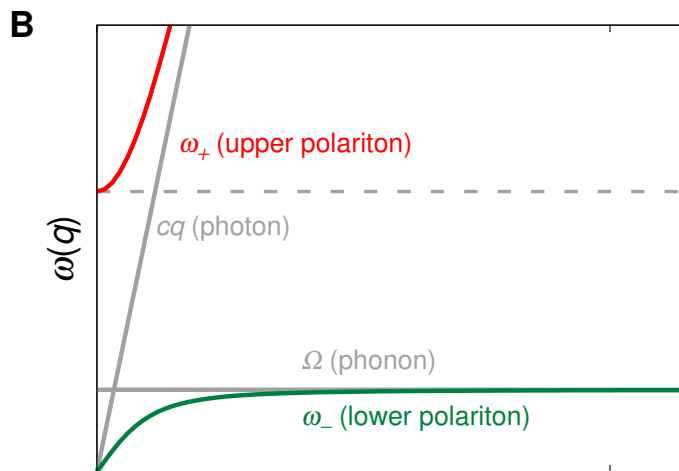


$$\hat{\Sigma}(\vec{k}, i\omega_n) = i\omega_n [1 - Z(\vec{k}, i\omega_n)] \hat{\tau}_0 + \chi(\vec{k}, i\omega_n) \hat{\tau}_3 + \phi(\vec{k}, i\omega_n) \hat{\tau}_1$$

$$\lambda \equiv Z(\vec{k}_F, i\pi/\beta) - 1$$

Mass enhancement:  $m^*/m = 1 + \lambda$

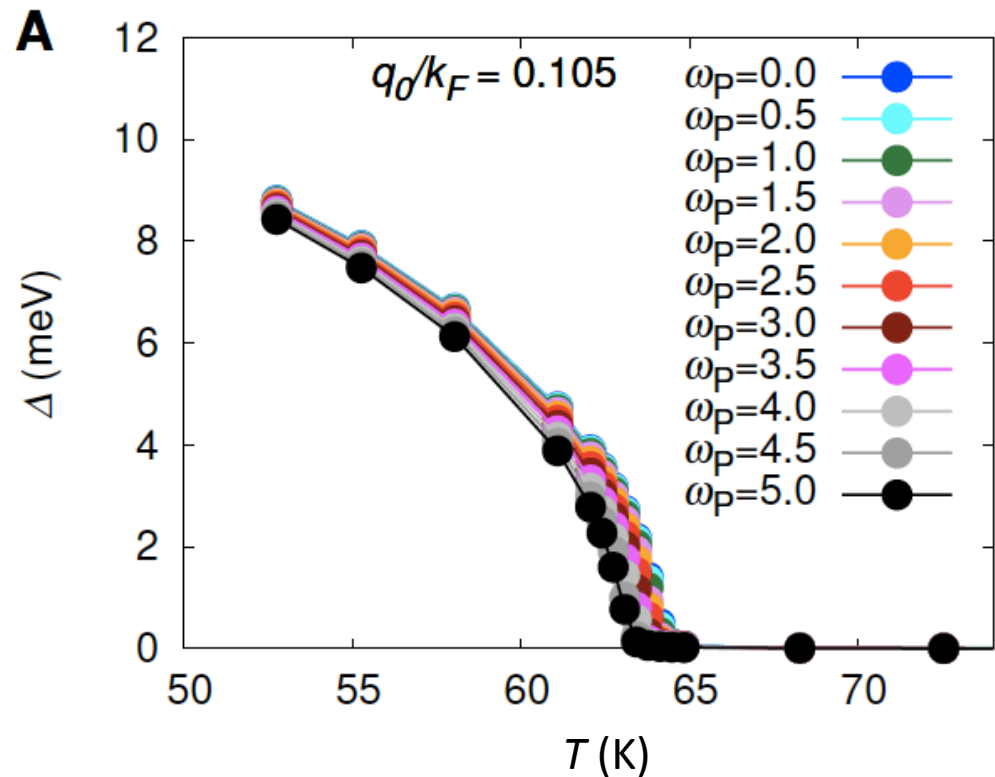
# Cavity materials: Phonon polaritons



enhanced electron-phonon coupling,  
controlled by cavity volume



# Cavity superconductivity?



suppressed superconductivity despite enhanced el-ph coupling

reason for suppression: forward scattering

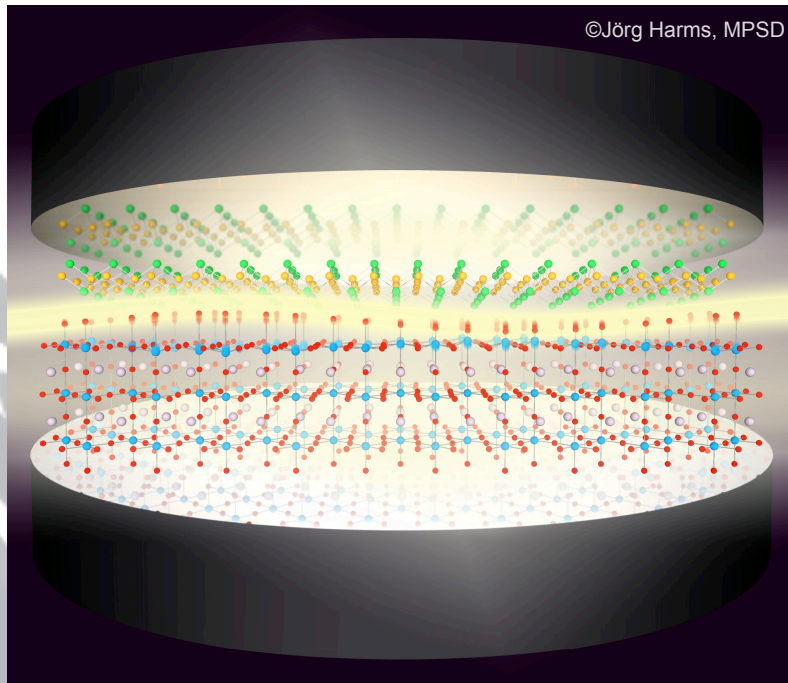
$$T_C \approx \frac{\lambda\Omega}{2 + 3\lambda}$$

vs.  $T_{C,BCS} \approx 1.13\Omega \exp(-\frac{1}{\lambda})$   
q-independent scattering

# Status as of October 2019 ...

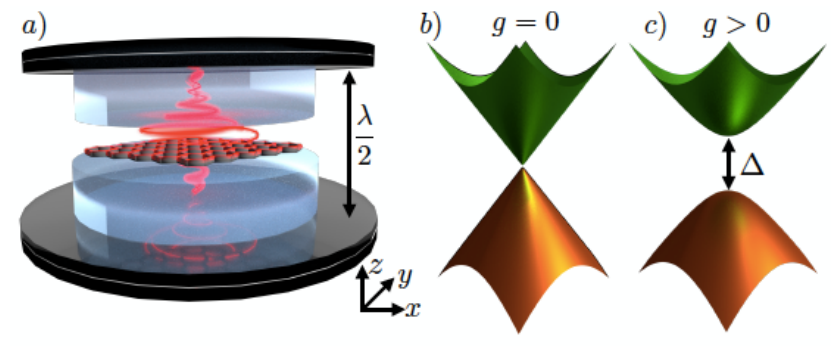
- cavity leads to **enhanced electron-phonon coupling**
- FeSe/STO: works in conjunction with other pairing mechanisms
- can one also enhance superconductivity?

*M. A. Sentef, M. Ruggenthaler, A. Rubio, arXiv:1802.09437  
Science Advances 4, eaau6969 (2018)*



Cavity-induced quantum-anomalous  
Hall effect in graphene:

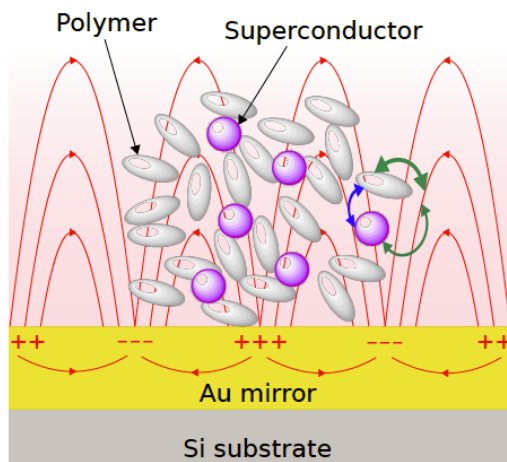
*Xiao Wang, E. Ronca, M. A. Sentef  
arXiv:1903.00339, PRB 2019*



# Exploring Superconductivity under Strong Coupling with the Vacuum Electromagnetic Field

A. Thomas<sup>1</sup>, E. Devaux<sup>1</sup>, K. Nagarajan<sup>1</sup>, T. Chervy<sup>1</sup>, M. Seidel<sup>1</sup>, D. Hagenmüller<sup>1</sup>, S. Schütz<sup>1</sup>,

J. Schachenmayer<sup>1</sup>, C. Genet<sup>1</sup>, G. Pupillo<sup>1\*</sup> & T. W. Ebbesen<sup>1\*</sup>



... consistent with enhanced electron-phonon coupling due to polariton formation and mode softening

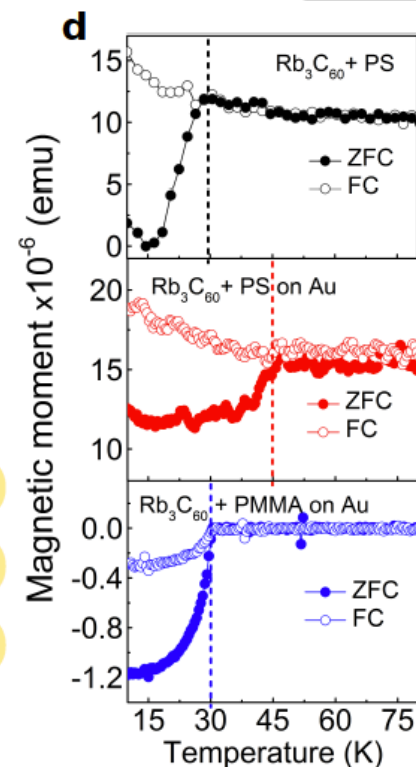
By placing the superconductor-surface plas-

mon system in a SQUID magnetometer, we find that the superconducting transition tem-

peratures ( $T_c$ ) for both compounds are modified in the absence of any external laser field.

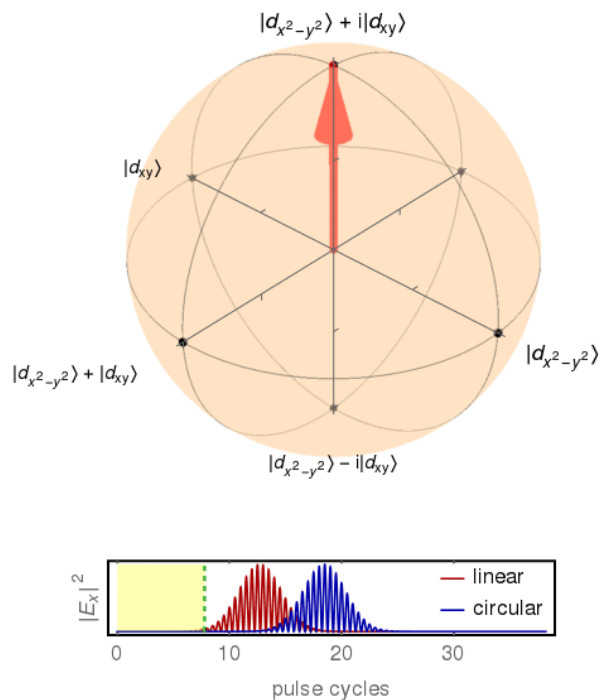
For YBCO,  $T_c$  decreases from 92 K to 86 K while for  $Rb_3C_{60}$ , it increases from 30 K to 45

K at normal pressures.

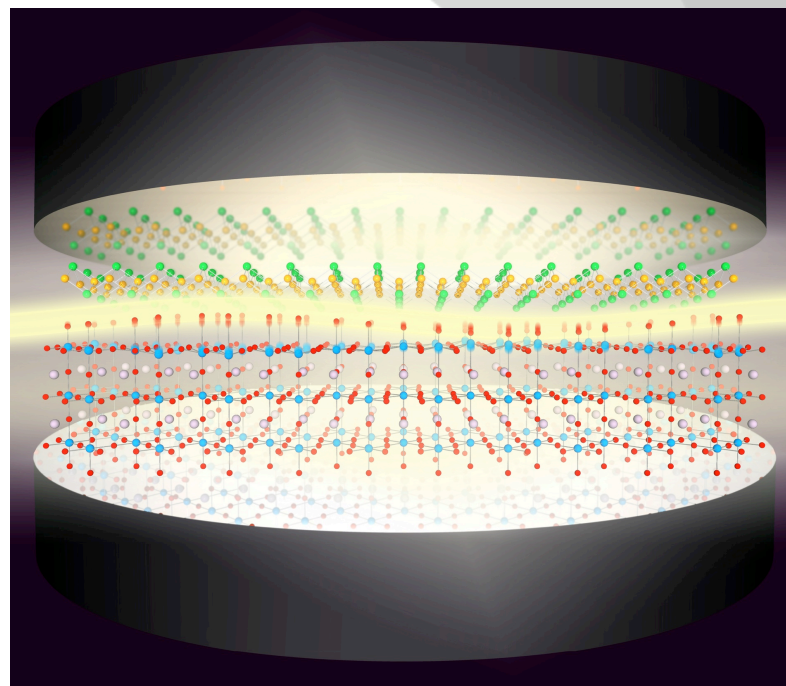


# Summary

*M. Claassen et al., Nat. Phys. 15, 766 (2019)*



*M. A. Sentef et al., Science Advances 4, eaau6969 (2018)*



Many opportunities for light-matter materials control!

Thank you for your attention!