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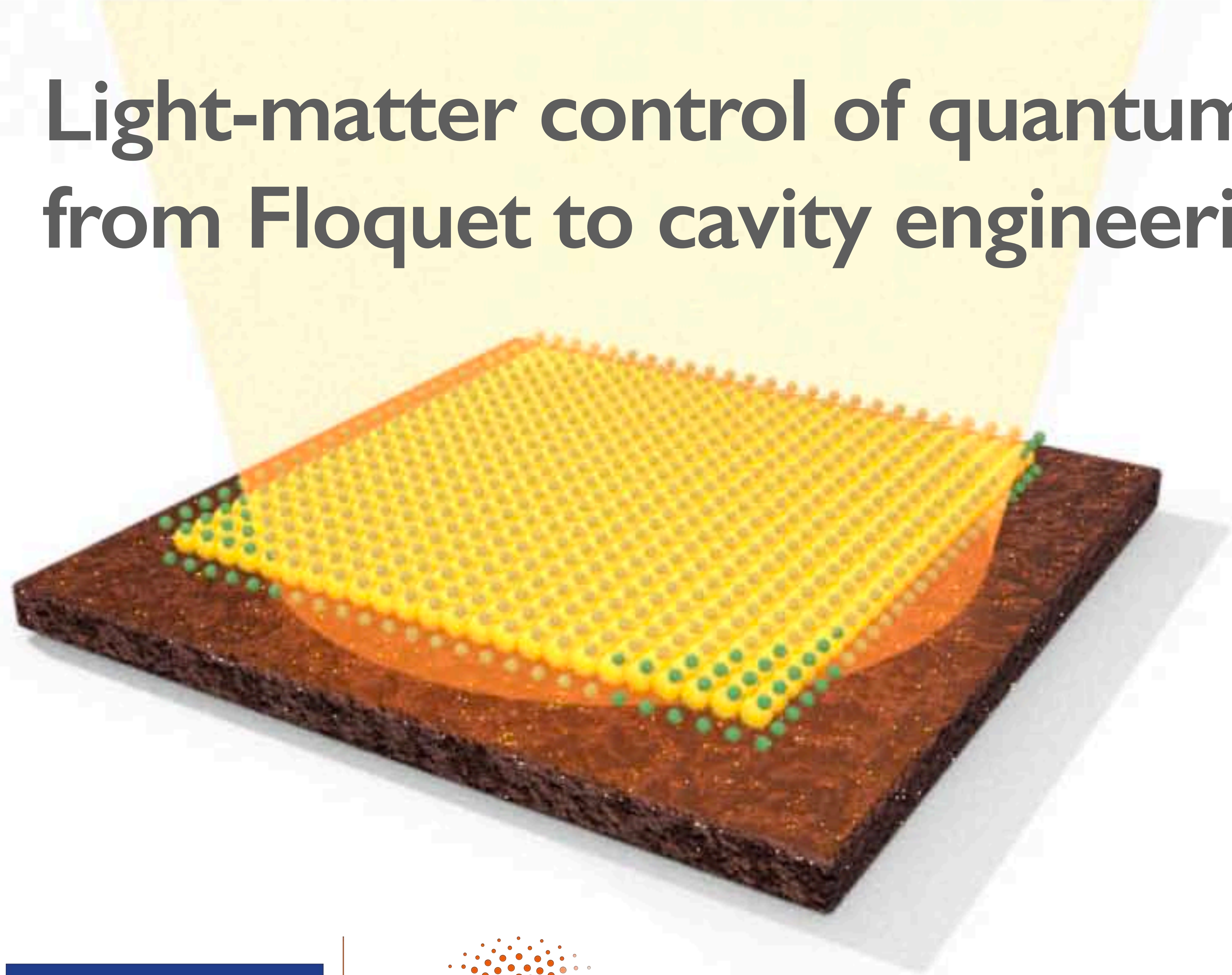
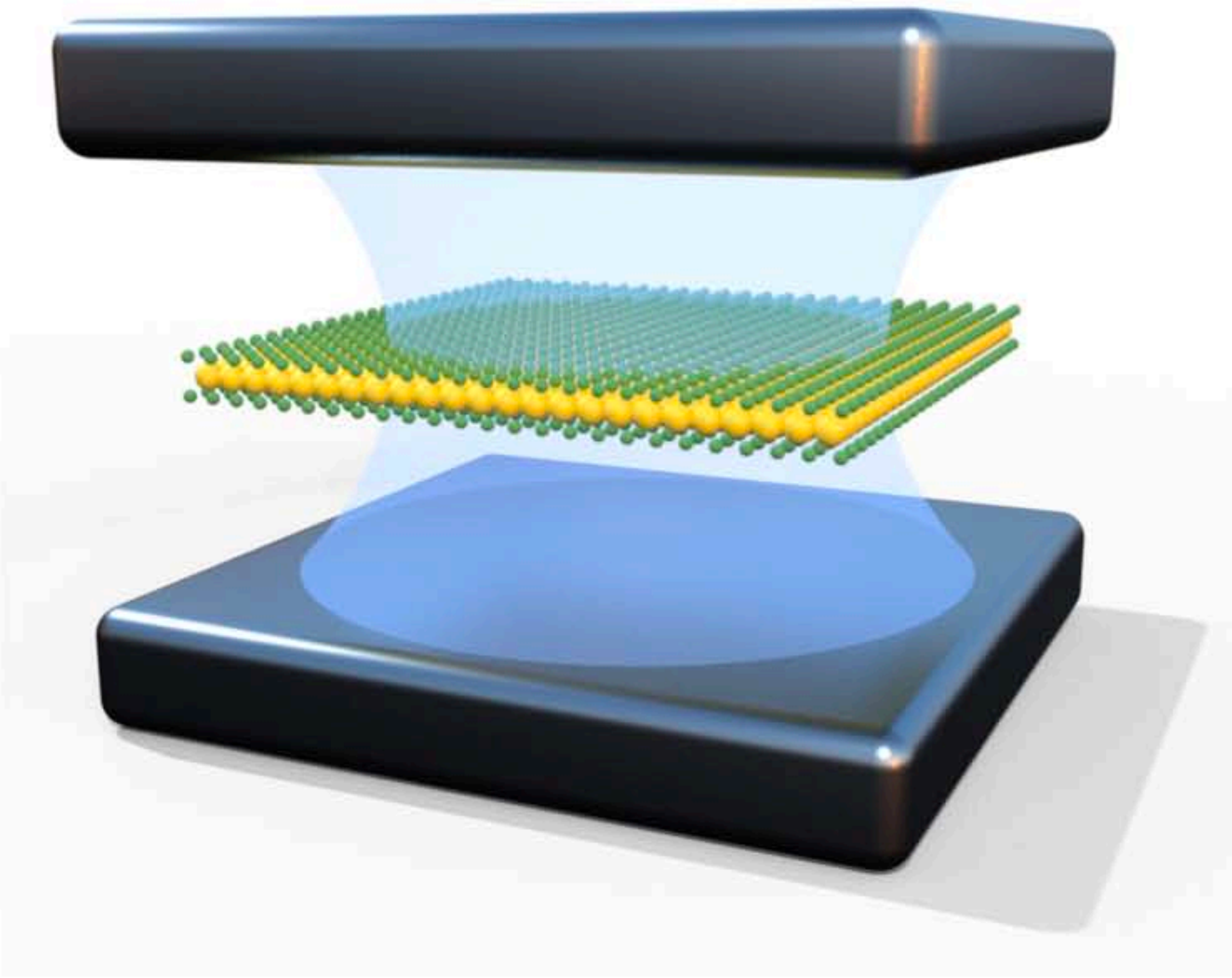
Michael A. Sentef

Light-matter control of quantum materials: from Floquet to cavity engineering





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IMPACT Cargèse, August 28, 2024

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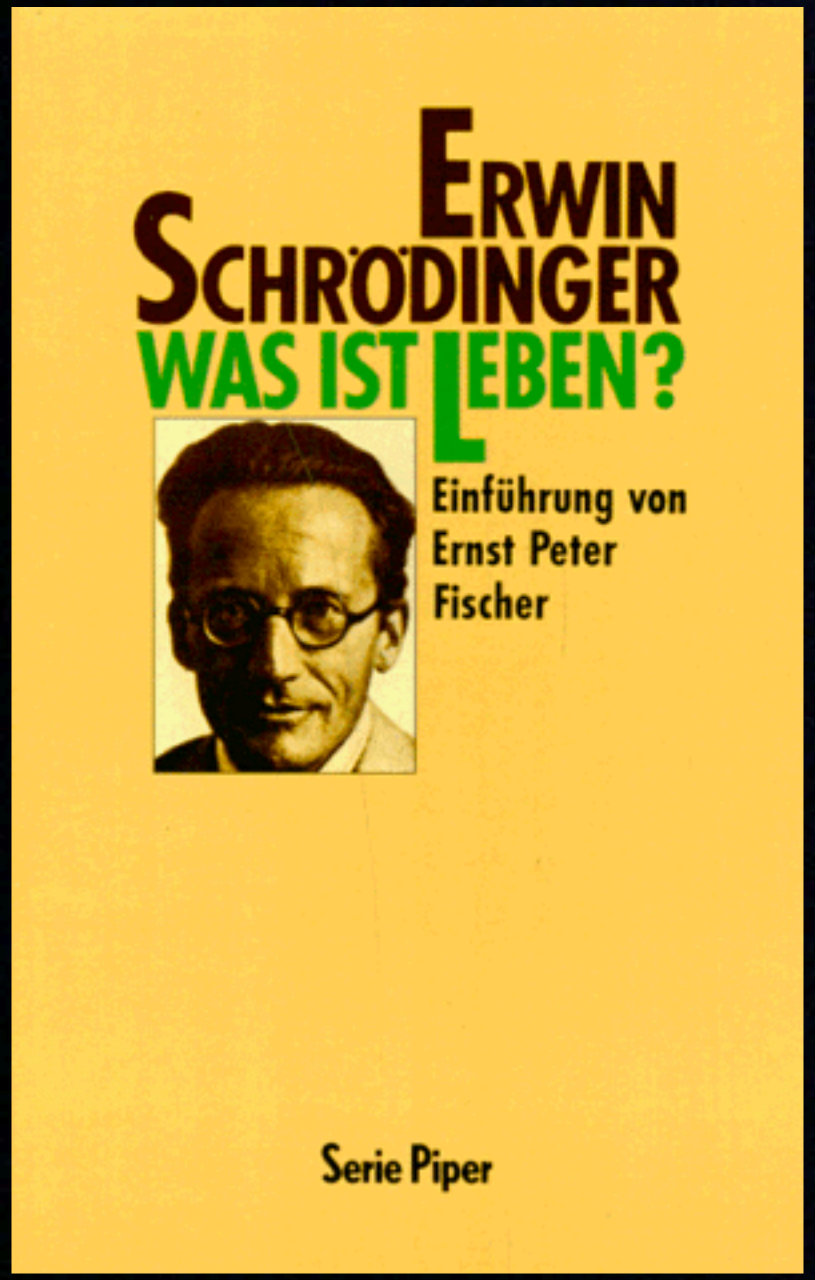
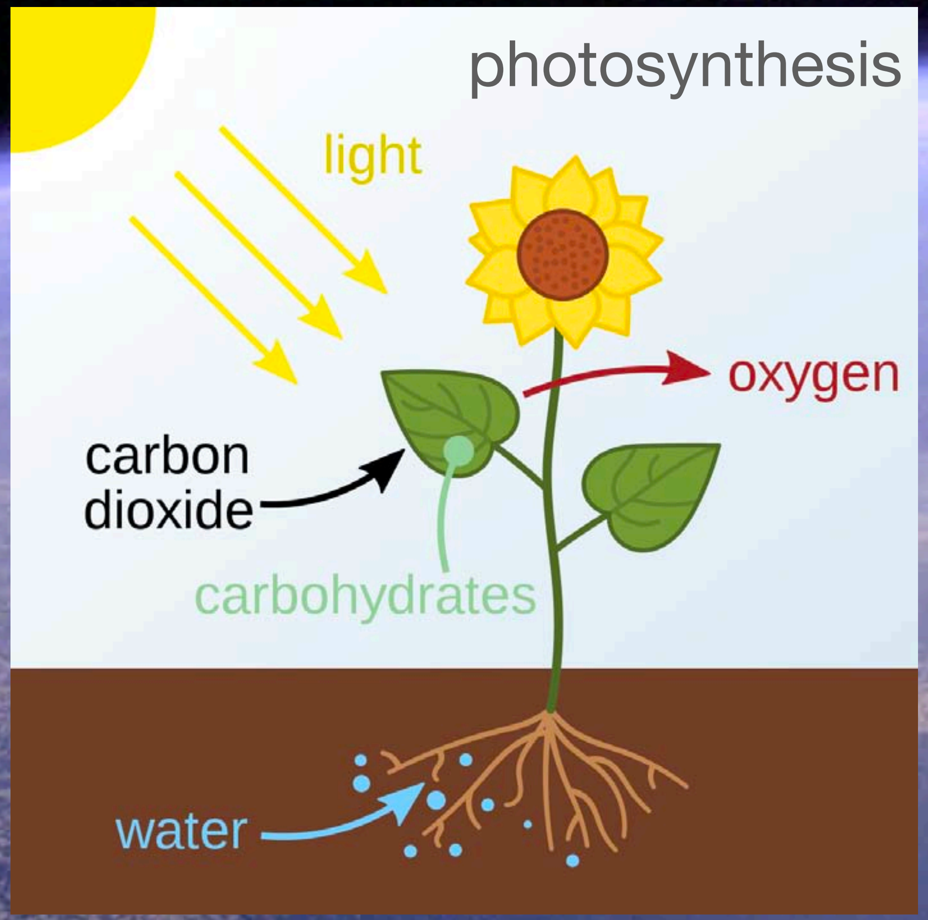
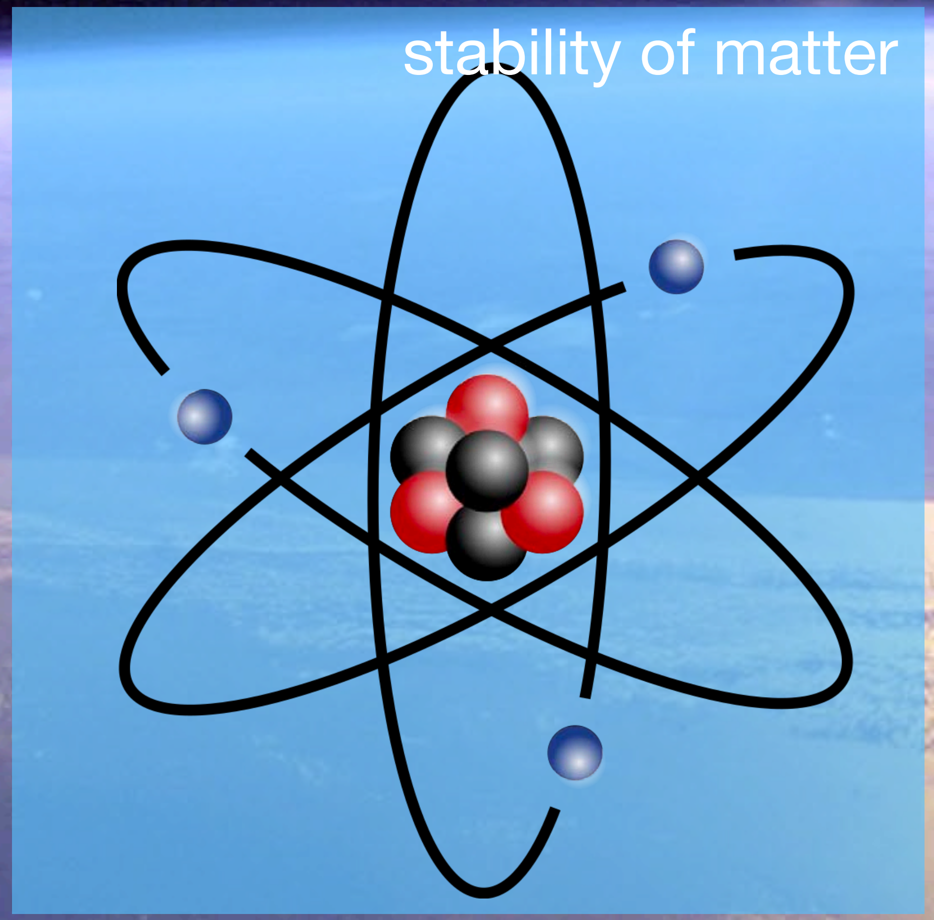
key importance of nonequilibrium for life



key importance of light-matter interactions for …





Can we employ light-matter interactions to change materials properties?

WIKIPEDIA
The Free Encyclopedia

Article Talk

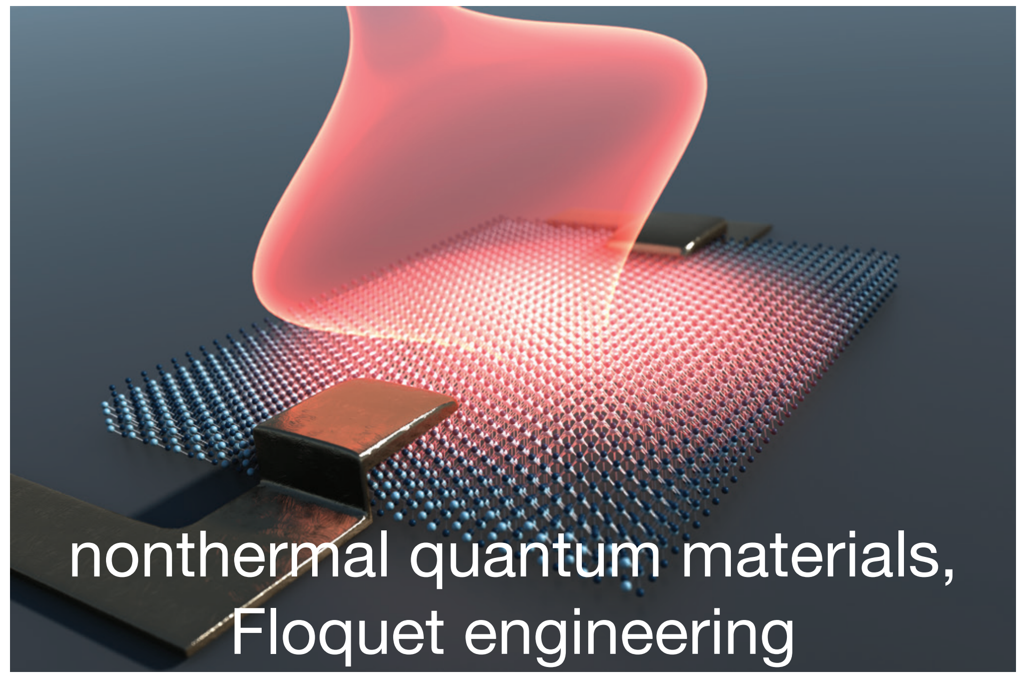
Fine-structure constant

From Wikipedia, the free encyclopedia

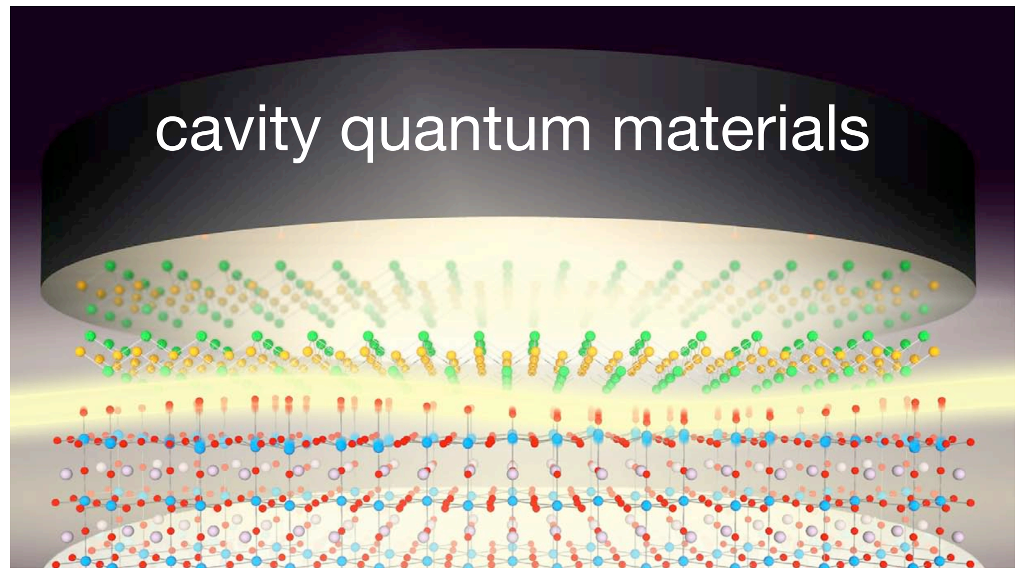
$$\alpha = \frac{1}{4\pi\varepsilon_0}\frac{e^2}{\hbar c} = \frac{\mu_0}{4\pi}\frac{e^2 c}{\hbar} = \frac{k_e e^2}{\hbar c} = \frac{e^2}{2\varepsilon_0 ch} = \frac{c\mu_0}{2R_K} = \frac{e^2 Z_0}{2h} = \frac{e^2 Z_0}{4\pi\hbar}$$

ing strength / photon number

strong laser driving



classical to q



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Colloquium: Nonthermal pathways to ultrafast control in quantum materials

Alberto de la Torre, Dante M. Kennes, Martin Claassen, Simon Gerber, James W. McIver, and Michael A. Sentef
Rev. Mod. Phys. **93**, 041002 – Published 14 October 2021

Home > Applied Physics Reviews > Volume 9, Issue 1 > 10.1063/5.0083825

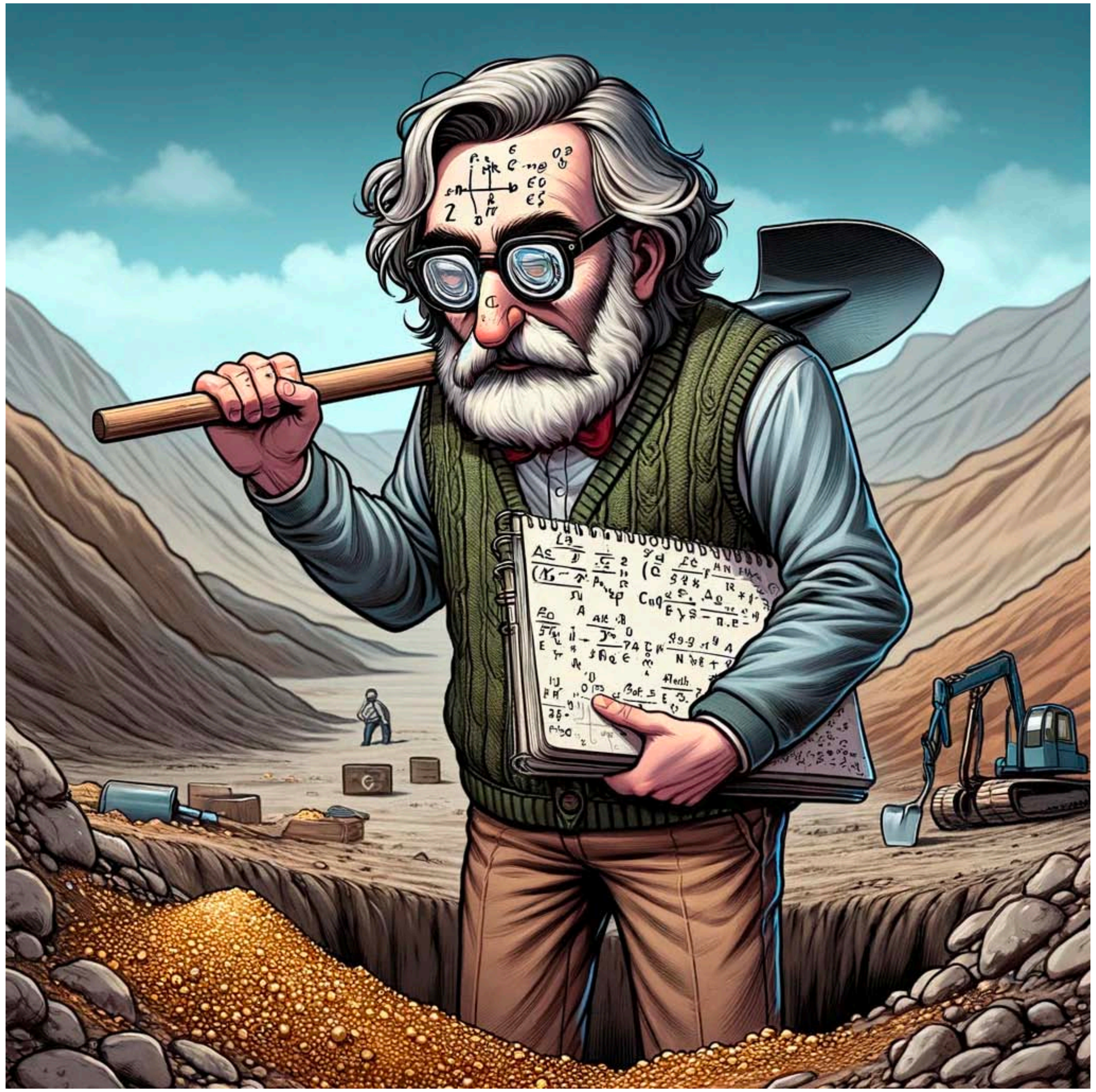
Open • Submitted: 30 December 2021 • Accepted: 31 January 2022 • Published Online: 25 February 2022

Cavity quantum materials F

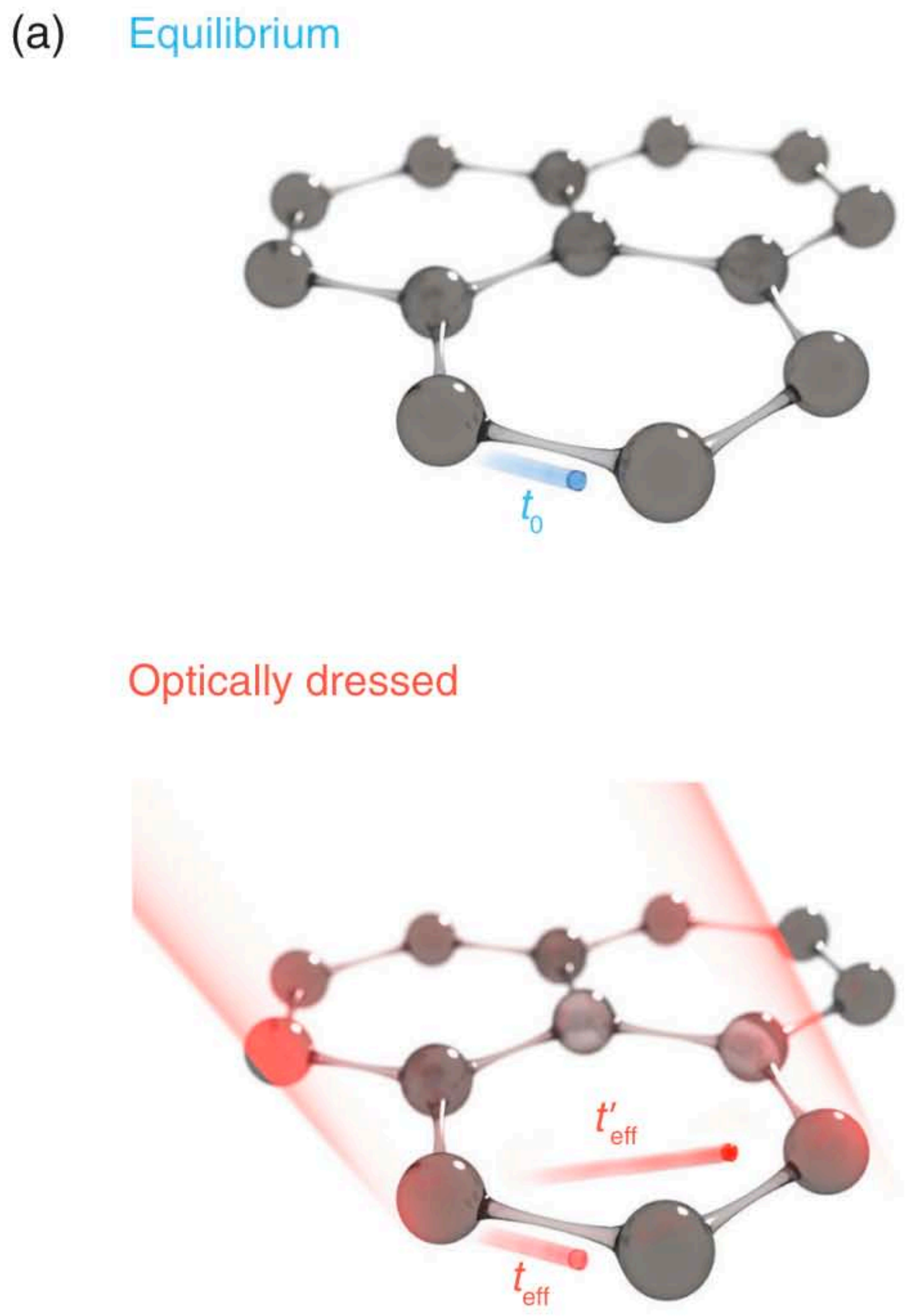
Applied Physics Reviews **9**, 011312 (2022); https://doi.org/10.1063/5.0083825

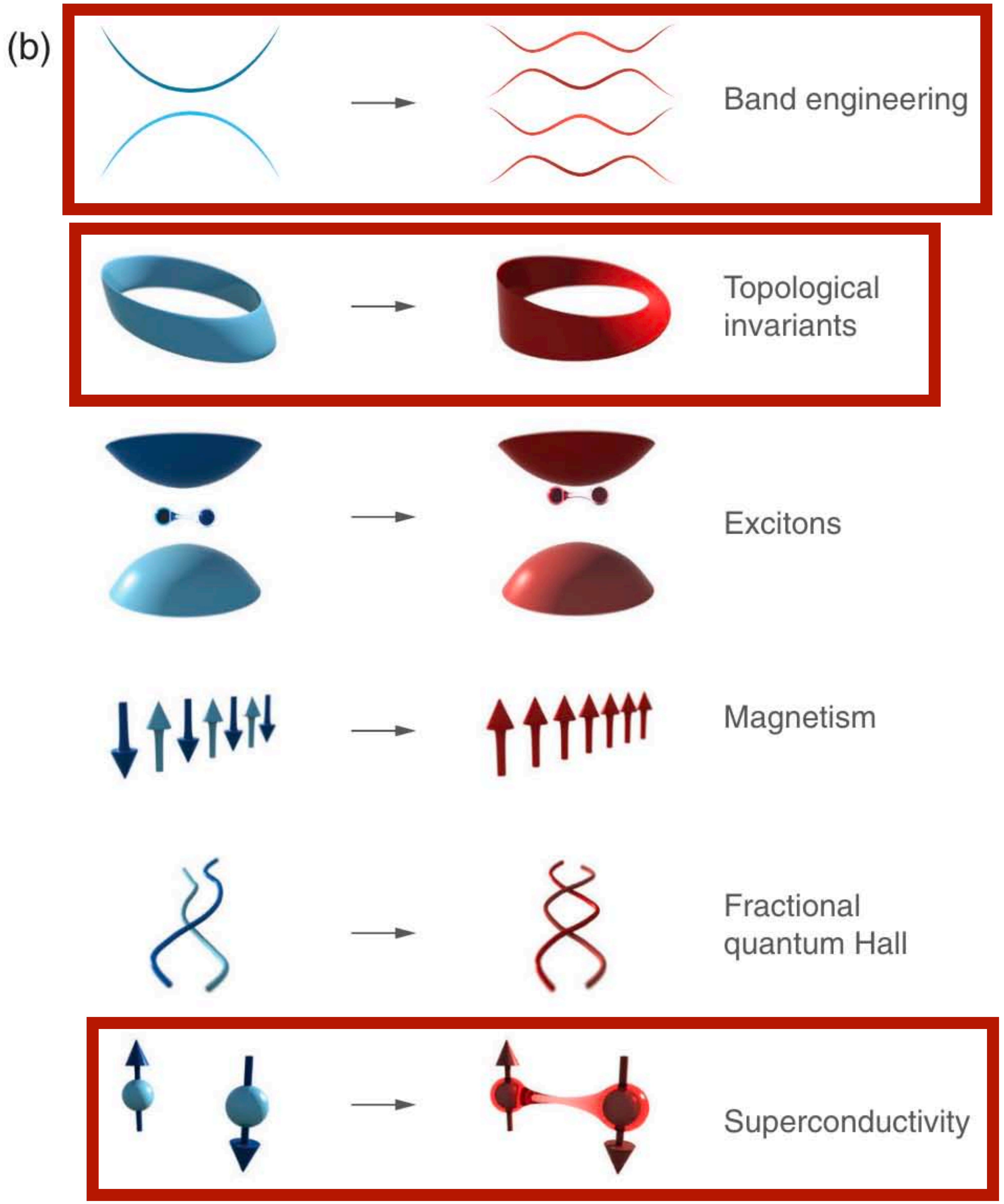
F. Schlawin[1,2], D. M. Kennes[1,3], *and* M. A. Sentef[1,a)]

Controlling quantum materials — gold rush



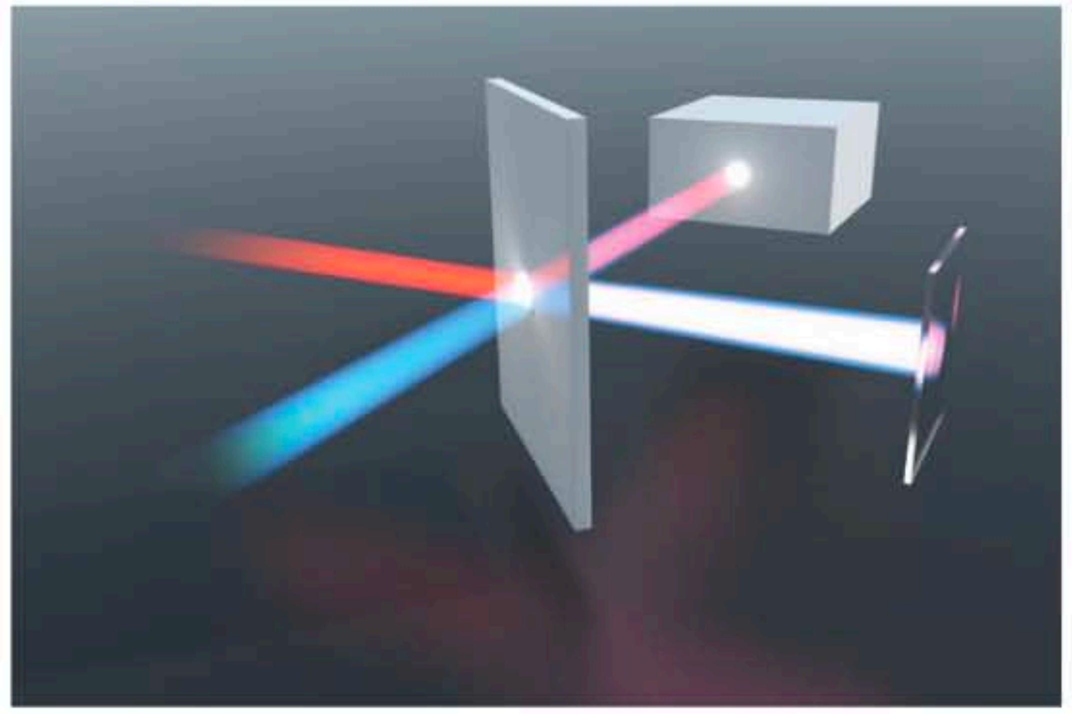
Controlling quantum materials — how?





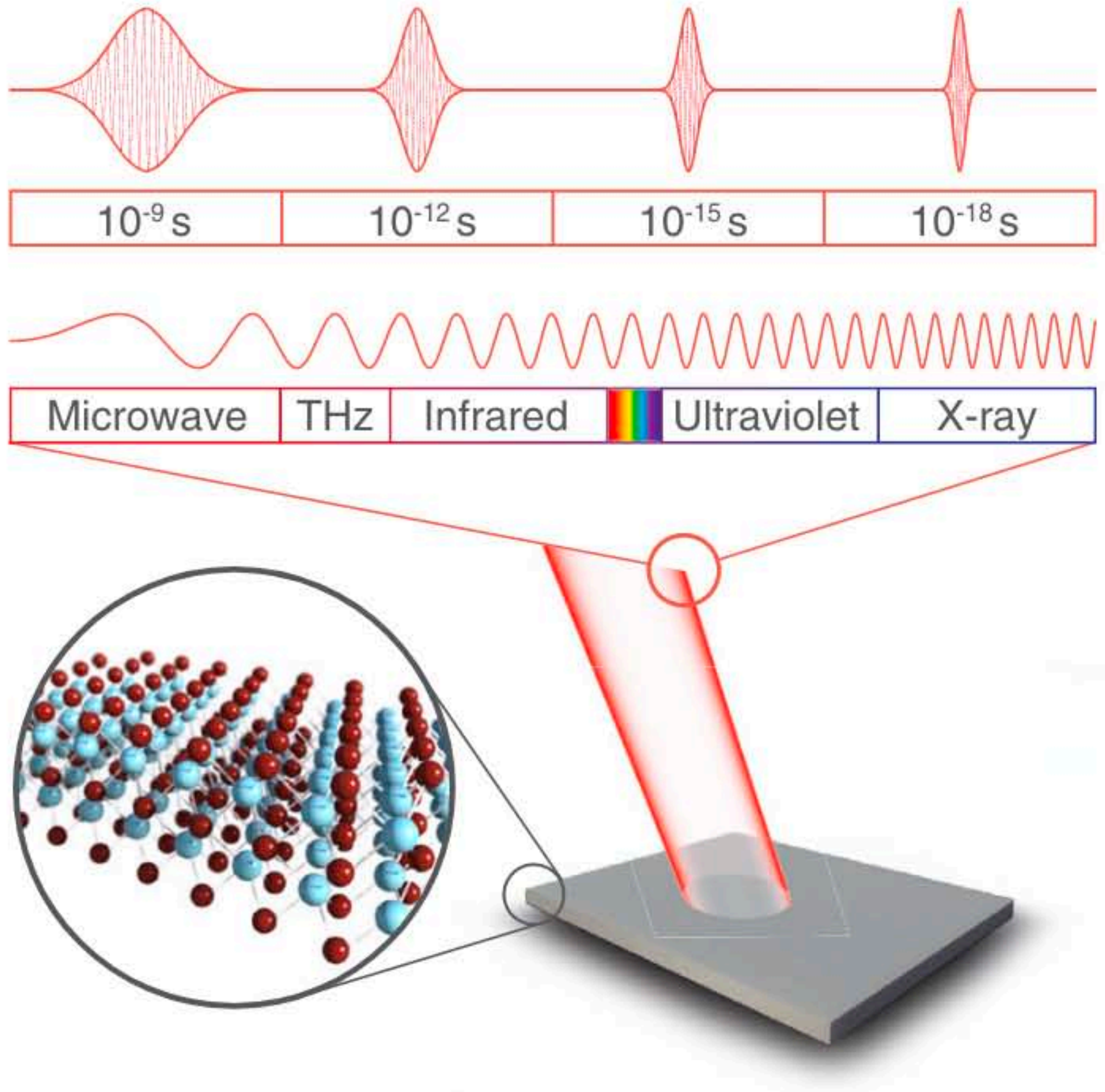
A. de la Torre et al.,
Rev. Mod. Phys. 93, 041002 (2021)

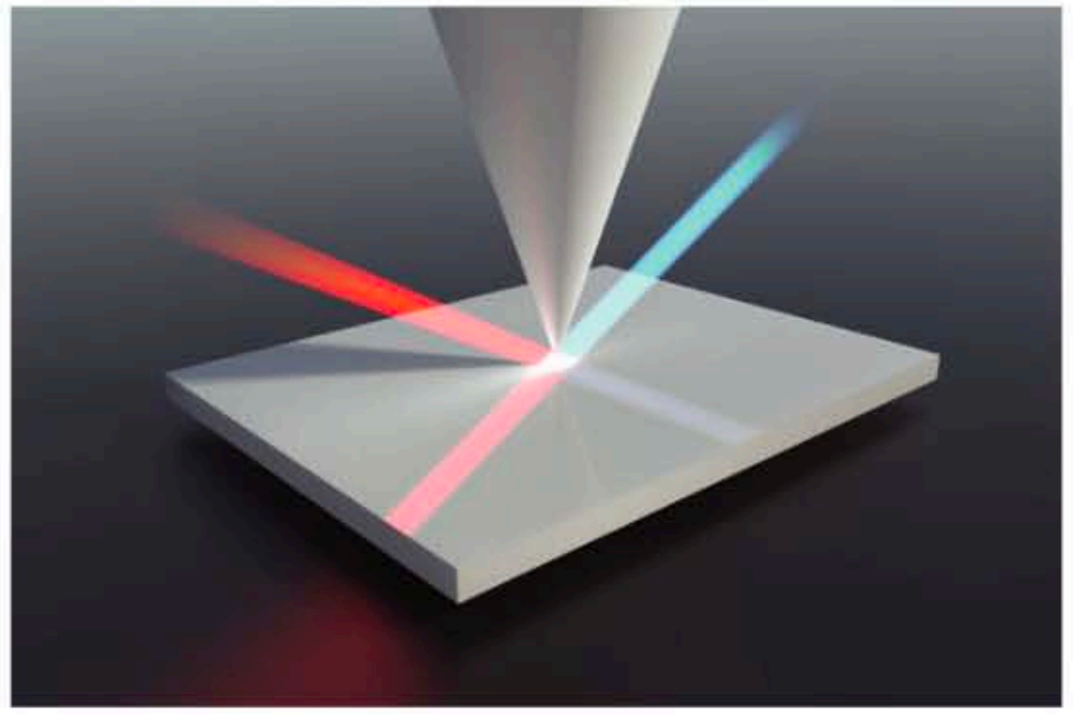
Dynamically establishing quantum correlations rather than randomizing an electronic system.



Optical probes

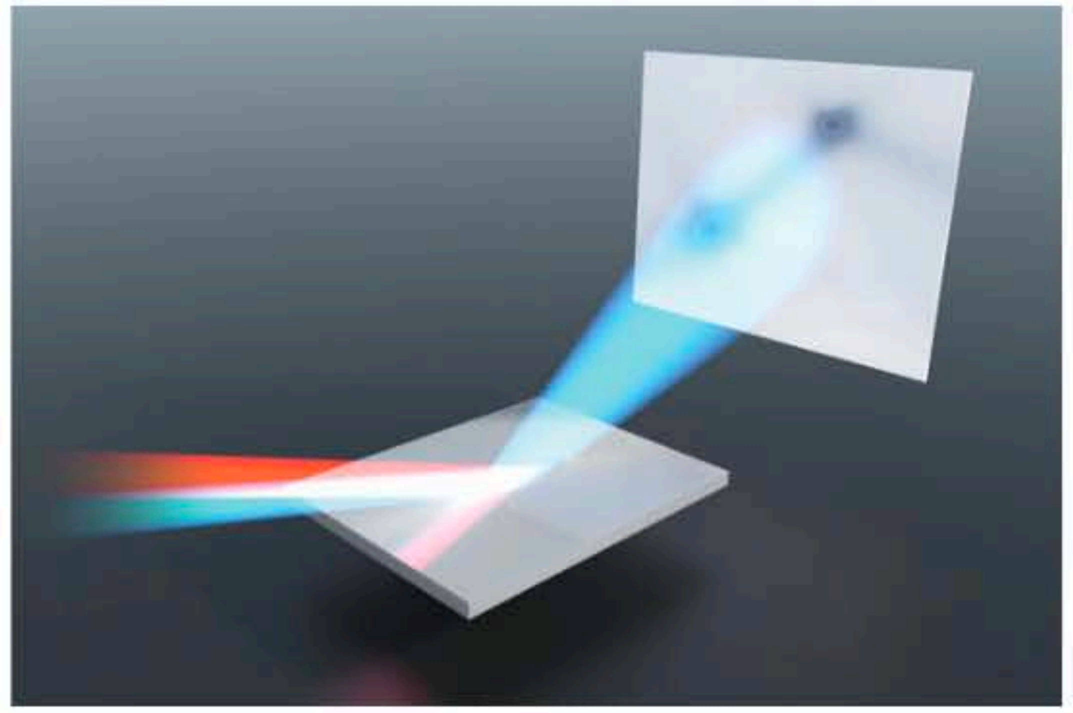
- Probes dielectric properties
- Flexible in implementation (spectral range, detection scheme, environment)
- fs time resolution





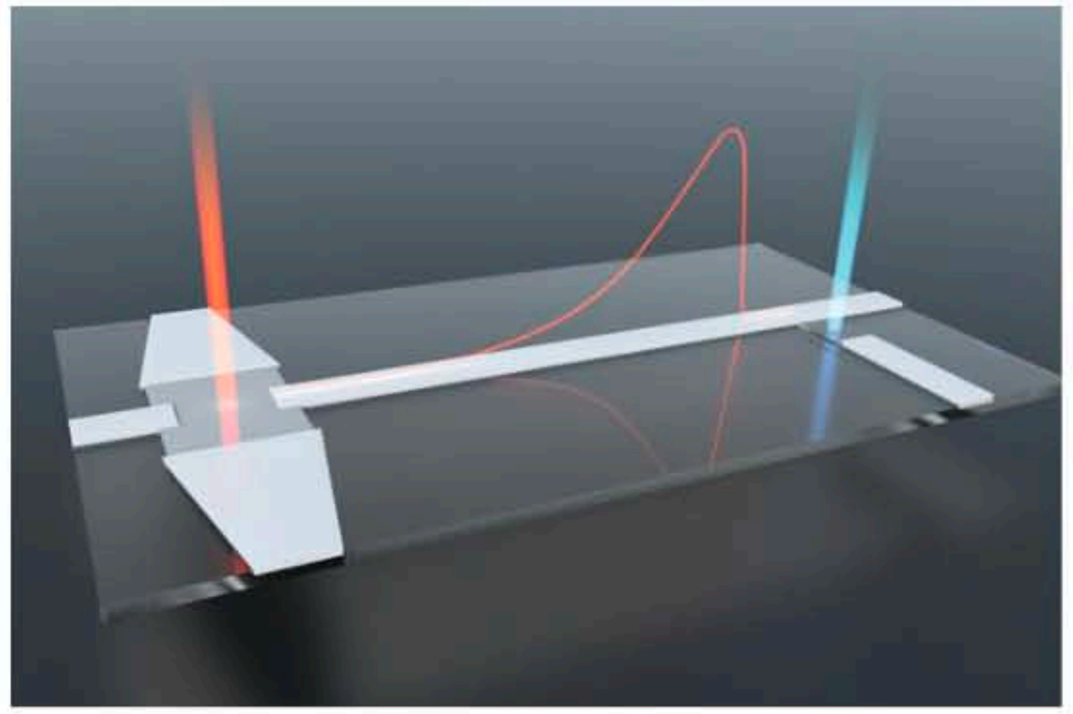
Scanning probes

- Probes optical constants in near-field (SNOM) or tunneling currents (STM)
- fs time resolution
- nm spatial resolution



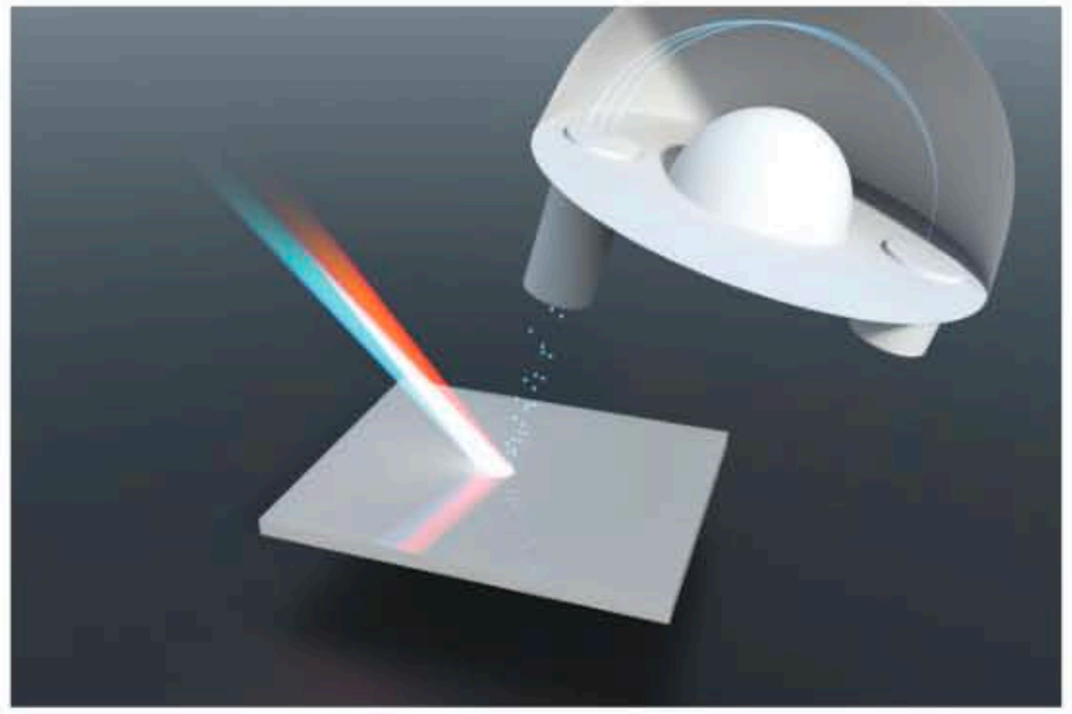
Scattering probes

- Probes structural dynamics and dynamics of electronic degrees of freedom at elemental resonances
- Access to dispersion relations via finite momentum transfer
- fs time resolution



Transport

- Probes transient photoconductivity
- Integrates well into microstructured devices
- Sub-ps time resolution



ARPES

- Probes time- and momentum-resolved carrier dynamics, and the evolution of electronic spectral functions
- Direct probe of electronic temperature
- Tunability of energy vs. time resolution (down to ~15 meV, ~30 fs)

A. de la Torre et al., Rev. Mod. Phys. 93, 041002 (2021)

also recommended: F. Boschini et al., Rev. Mod. Phys. 96, 015003 (2024)

+ need for theoretical/computational schemes to predict/understand/guide

Outline

Floquet basics
Floquet in solids
Floquet to cavity

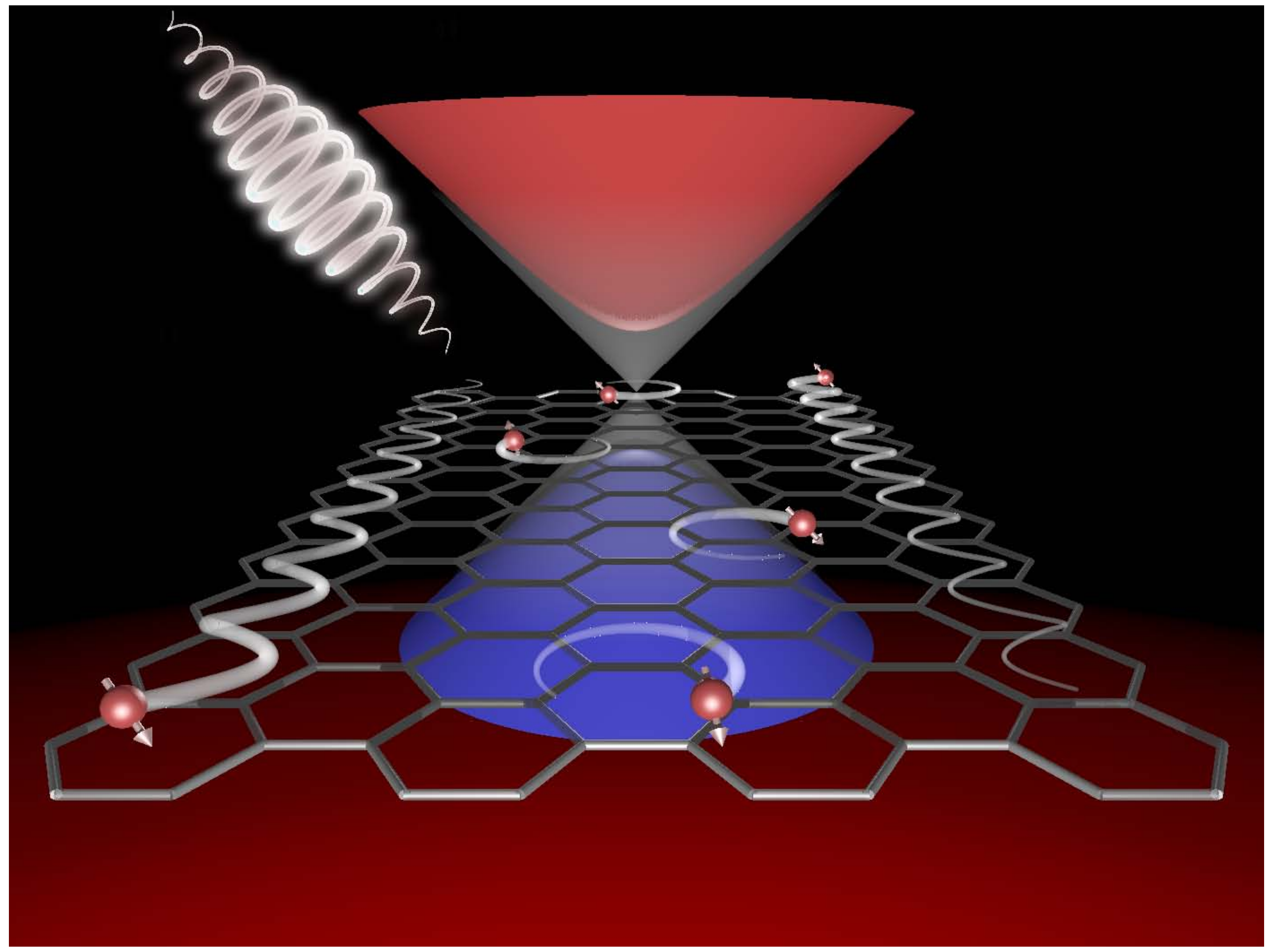
Outline

Floquet basics
Floquet in solids
Floquet to cavity

Example: Can one engineer the Haldane model dynamically?

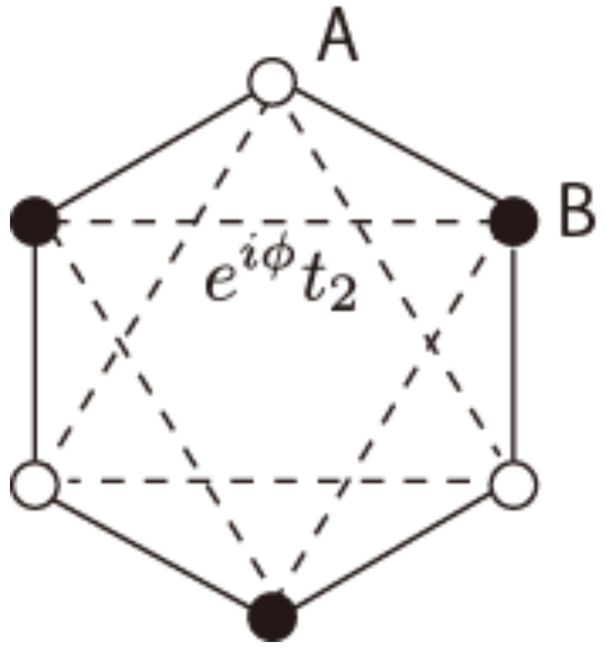
Quantum Hall effect without a magnetic field

Graphene + circularly polarized light
(breaks time reversal)

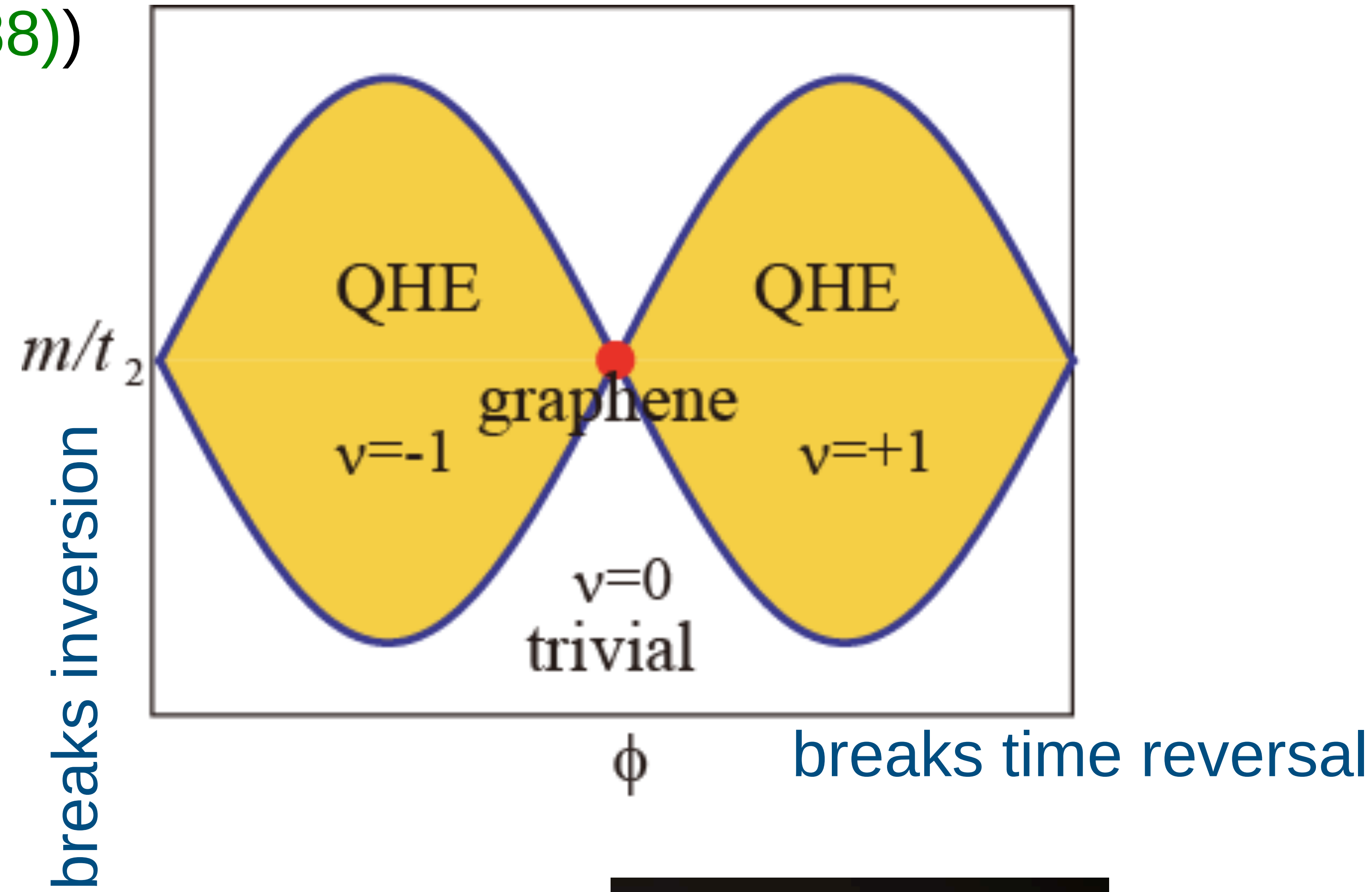


?

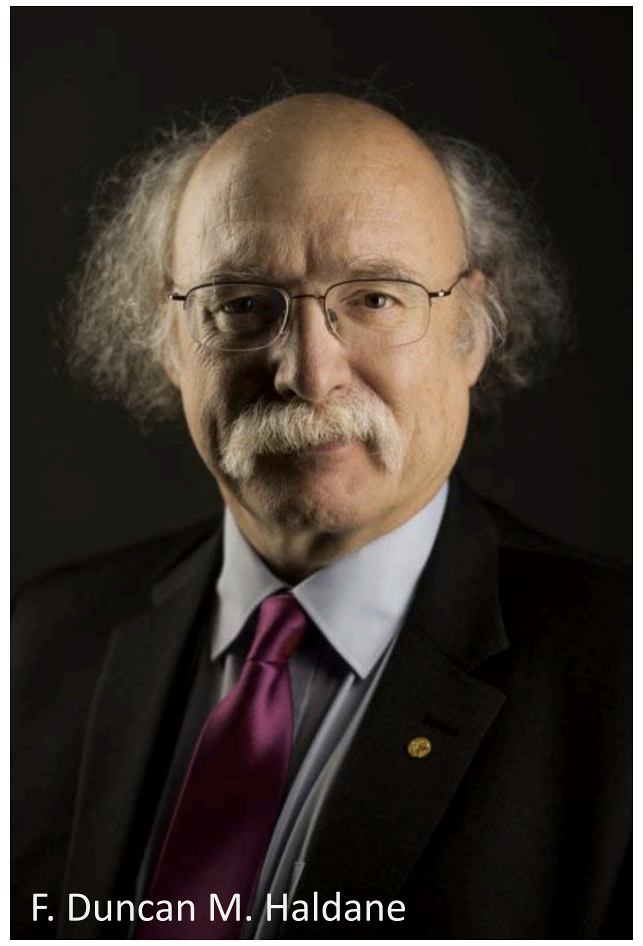
Haldane model (PRL 61, 2015 (1988))



Local flux ϕ
Staggered field $m = E_A - E_B$
Fictitious fields!

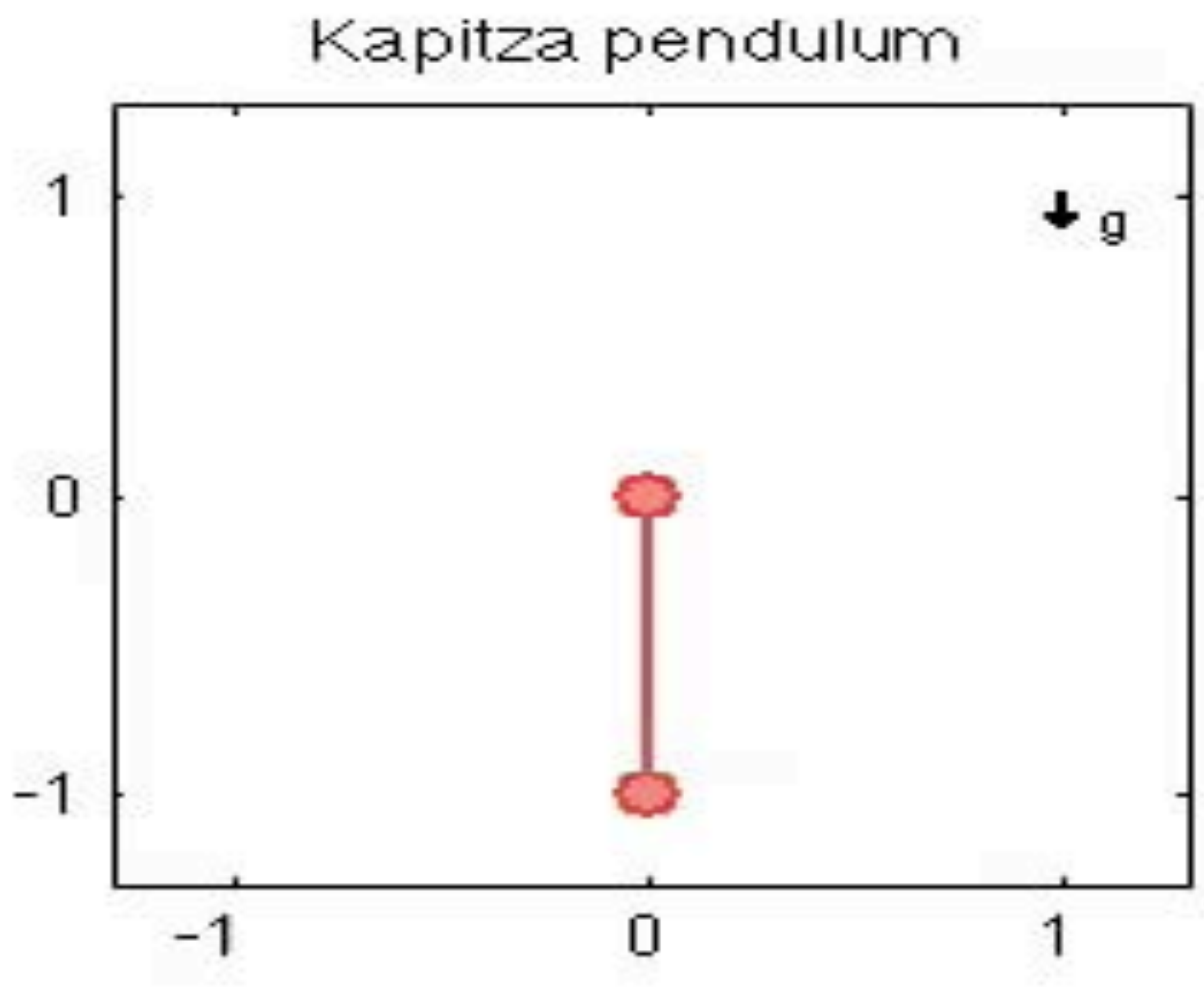


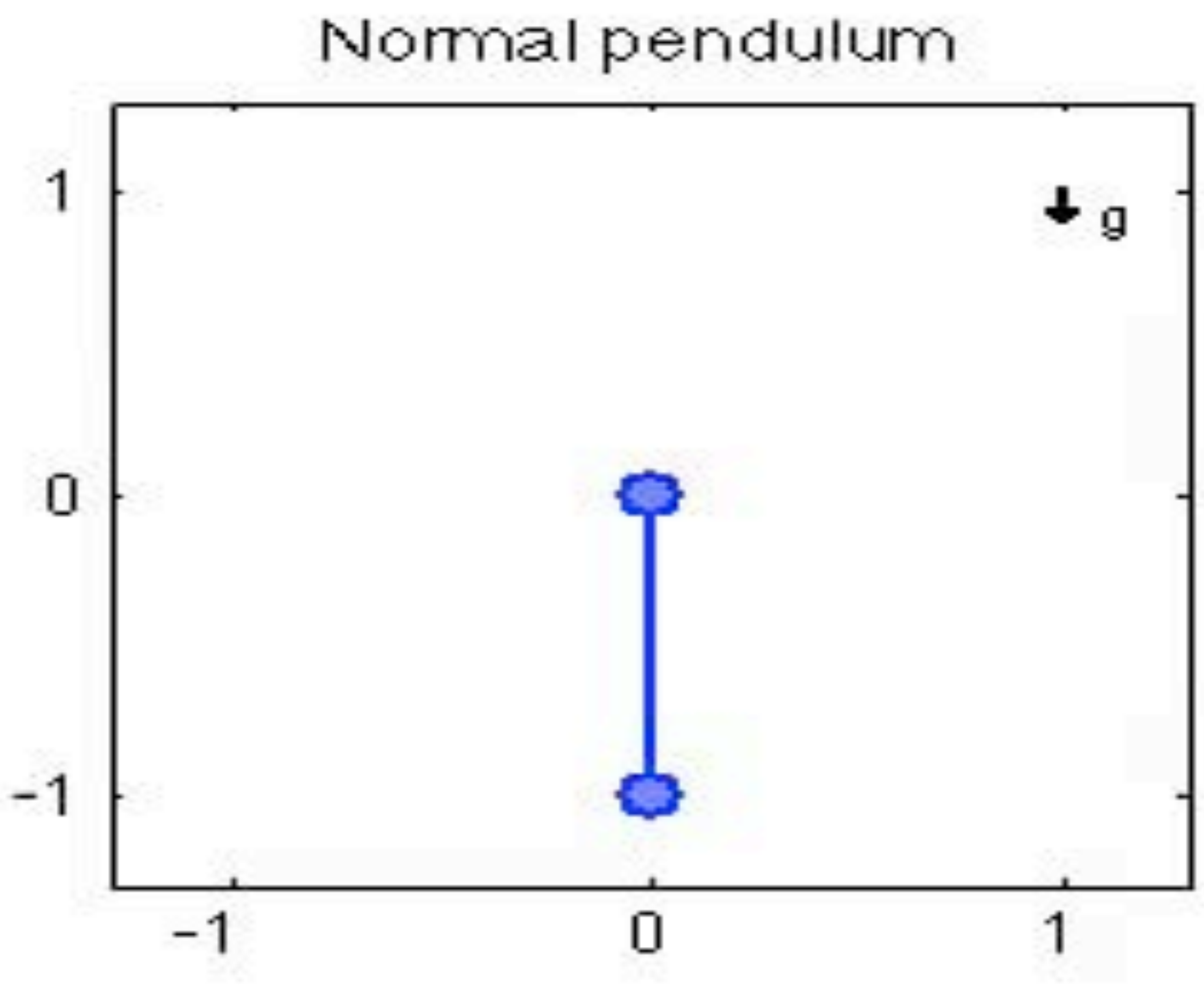
Need concept of Floquet engineering



(c) Nobel Media AB,
Photo: A. Mahmoud

Kapitza pendulum: dynamical stabilization of metastable state





Floquet theory in a nutshell

time periodic system

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

=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

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

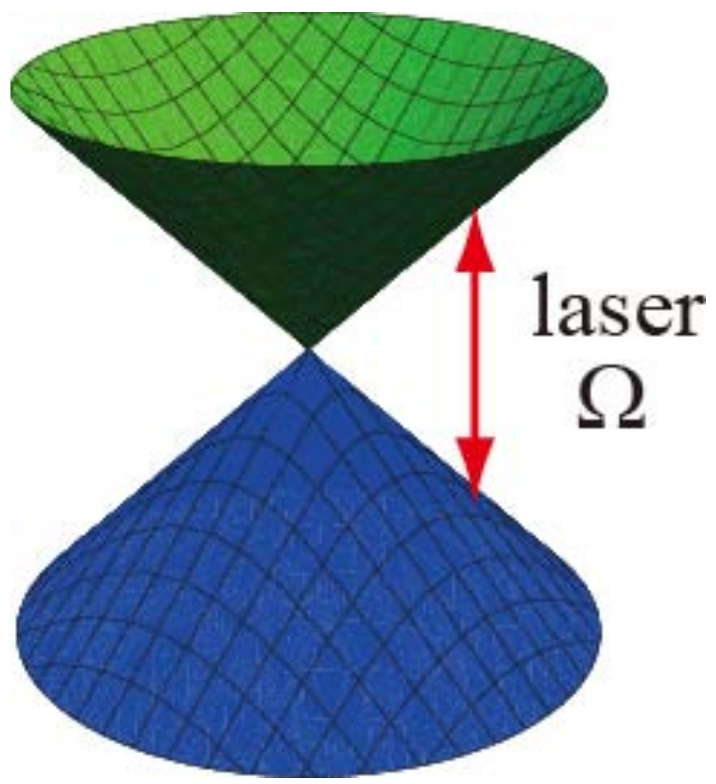
comes from the $i\partial_t$ term

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

~ absorption of *m* "photons"

Oka & Aoki, Phys. Rev. B 79, 081406(R) (2009)
Kitagawa et al., Phys. Rev. B 82, 235114 (2010)

Dirac fermion with circularly polarized laser



coupling to AC field

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

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

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

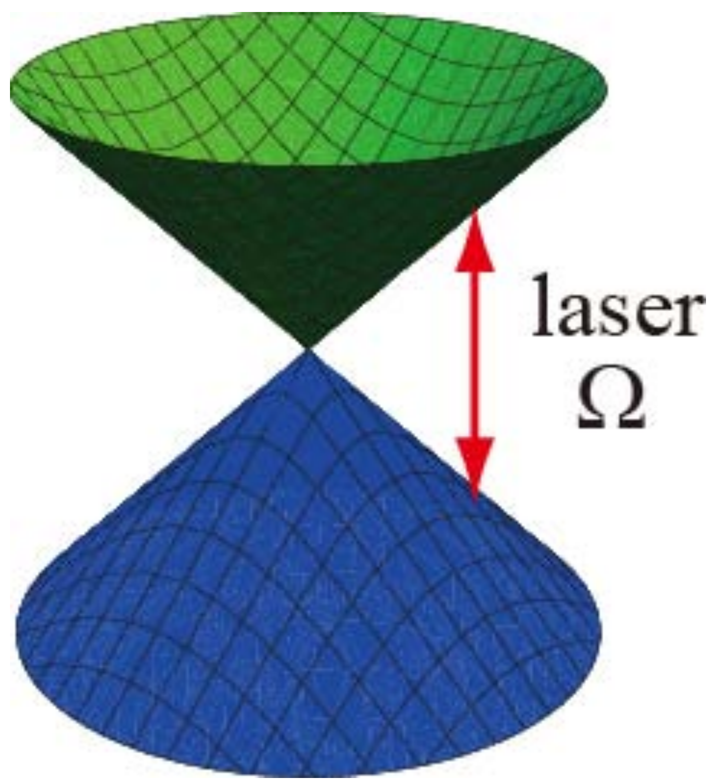
$$(\mathcal{H})^{mn} = \frac{1}{T}\int_0^T dt H(t) e^{i(m-n)\Omega t} + m\delta_{mn}\Omega I$$

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

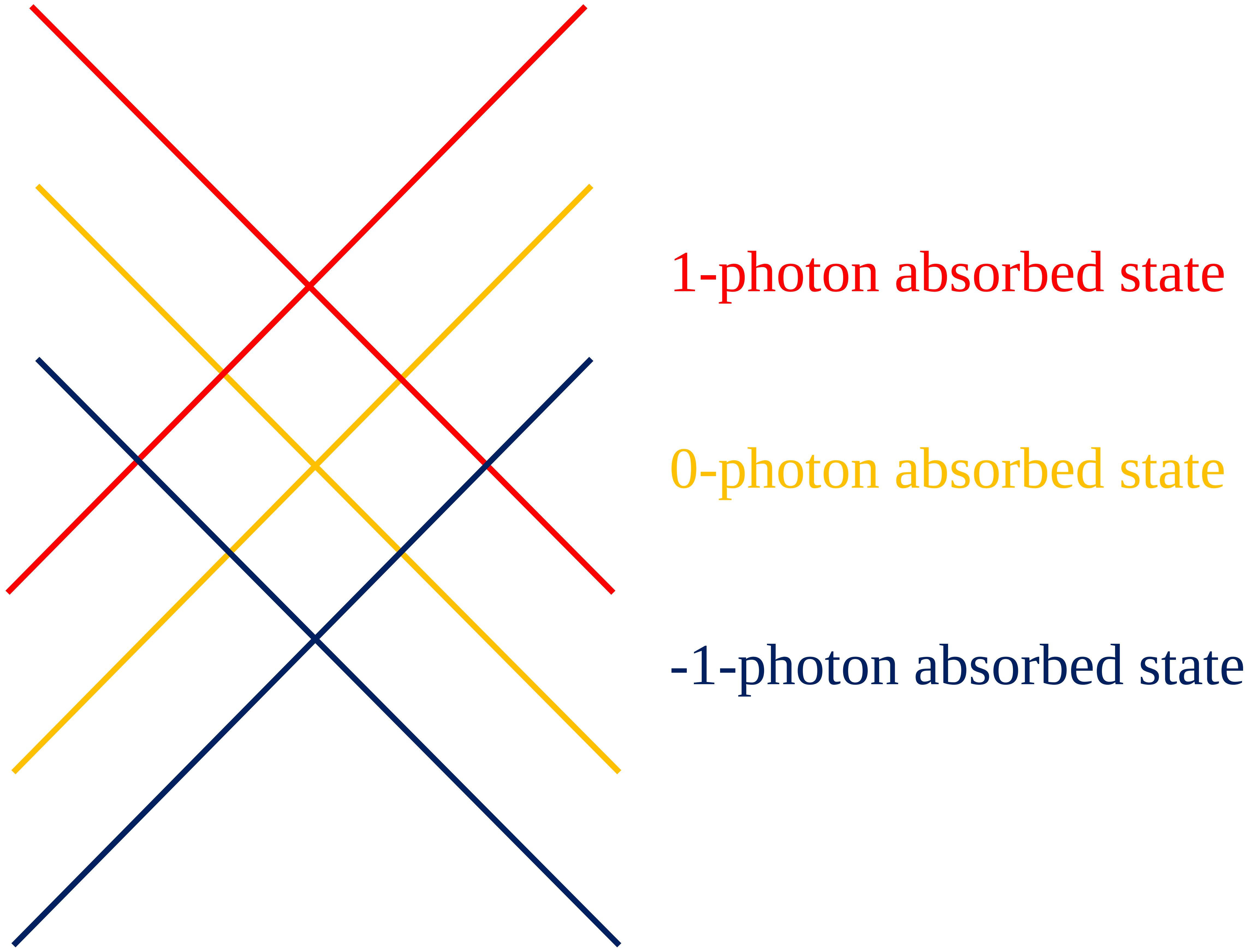
truncated at m=0,+1, -1 for display

Oka & Aoki, Phys. Rev. B 79, 081406(R) (2009)
Kitagawa et al., Phys. Rev. B 82, 235114 (2010)

Dirac fermion with circularly polarized laser

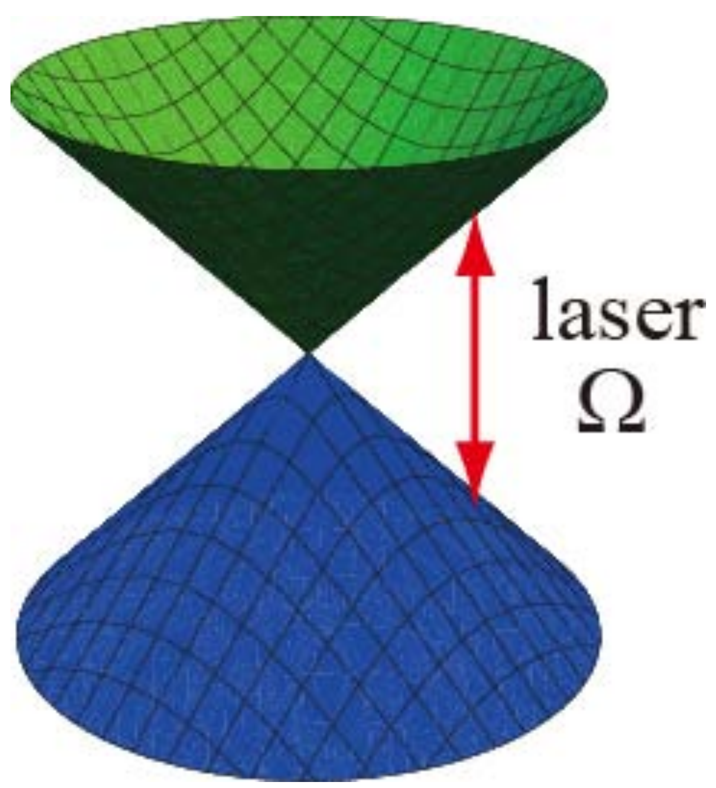


$$H^{\mathrm{Floquet}} = \begin{pmatrix} \Omega & k & 0 & A & 0 & 0 \\ \bar{k} & \Omega & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & k & 0 & A \\ A & 0 & \bar{k} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -\Omega & k \\ 0 & 0 & A & 0 & \bar{k} & -\Omega \end{pmatrix}$$

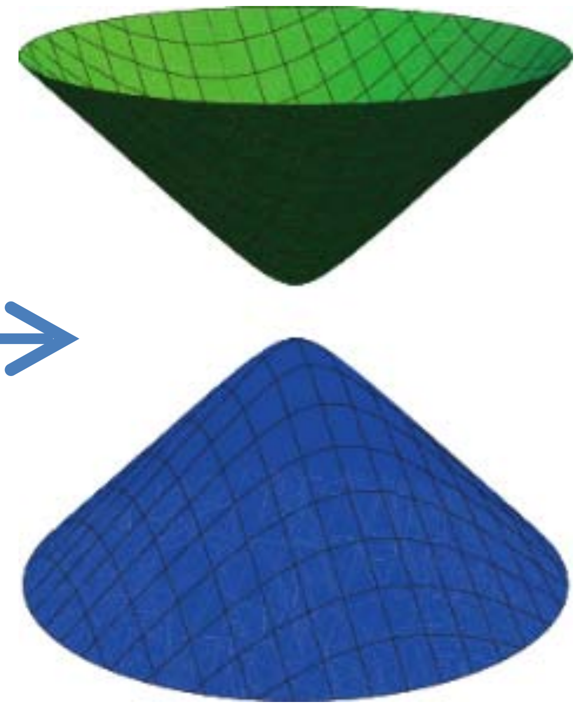


Oka & Aoki, Phys. Rev. B 79, 081406(R) (2009)
Kitagawa et al., Phys. Rev. B 82, 235114 (2010)

Dirac fermion with circularly polarized laser



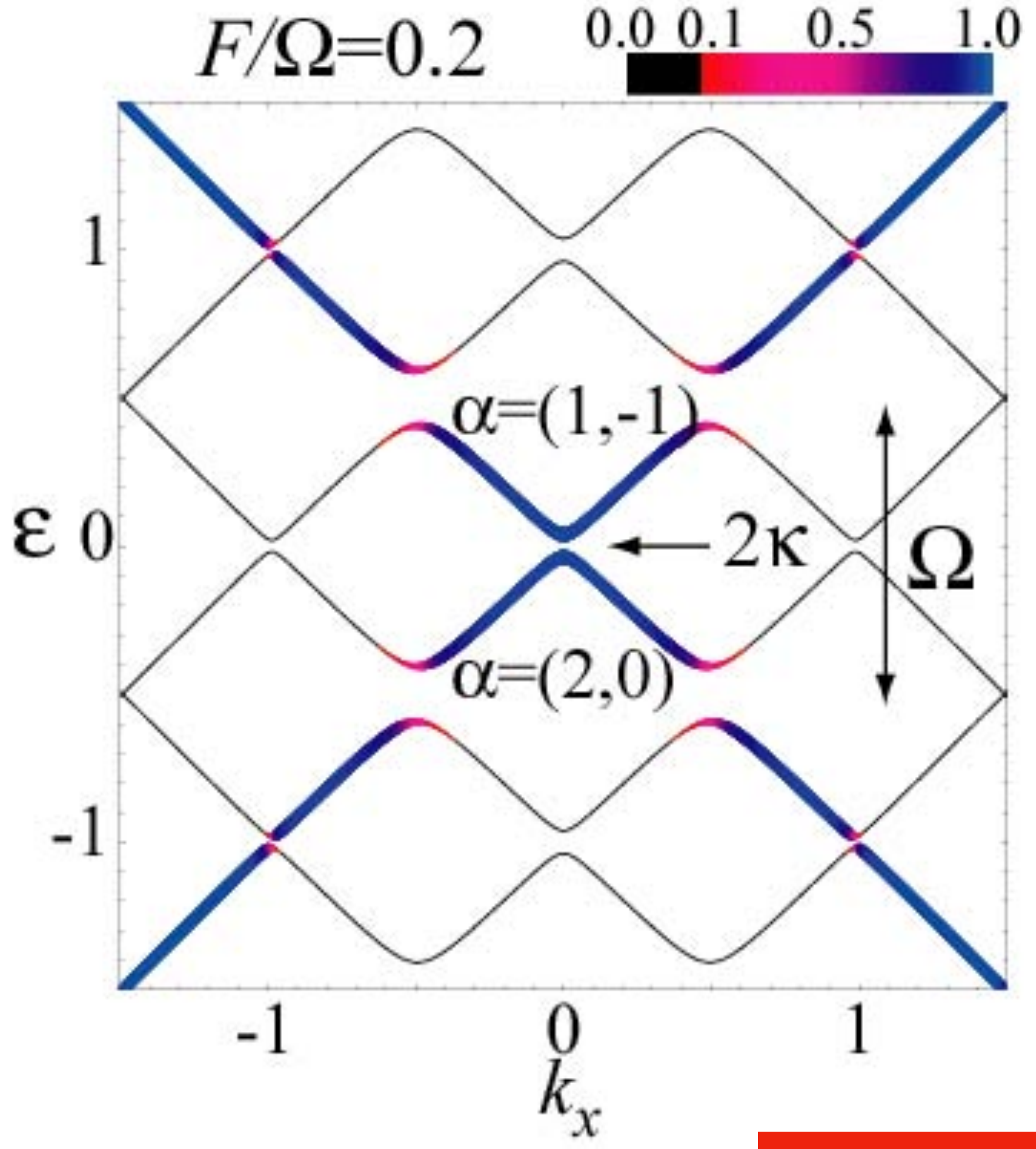
$$\kappa = \frac{\sqrt{4A^2+\Omega^2}-\Omega}{2} \sim A^2/\Omega$$



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

sign of mass term determined
by chirality of light

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



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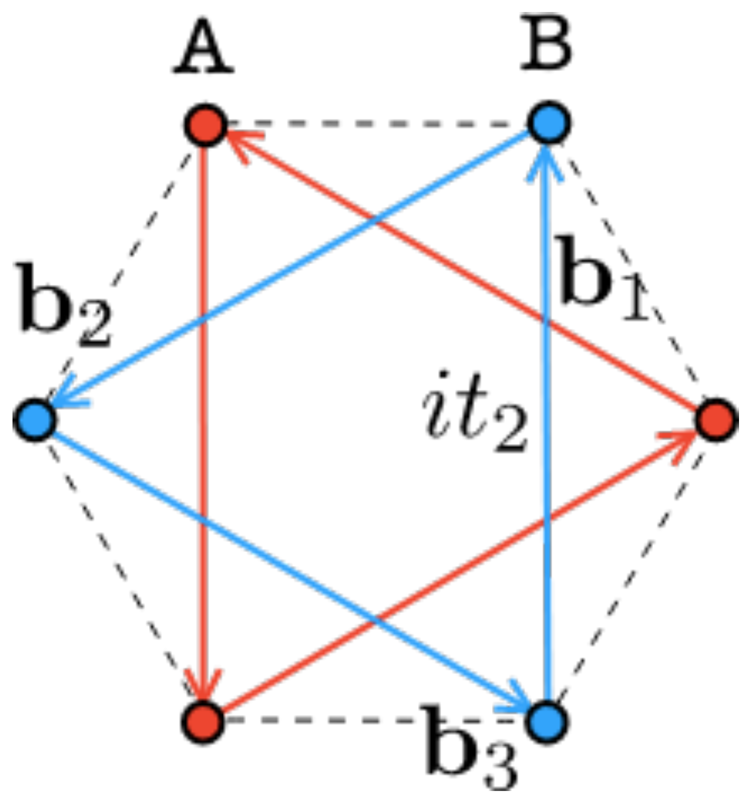
Haldane, PRL 61, 2015 (1988)



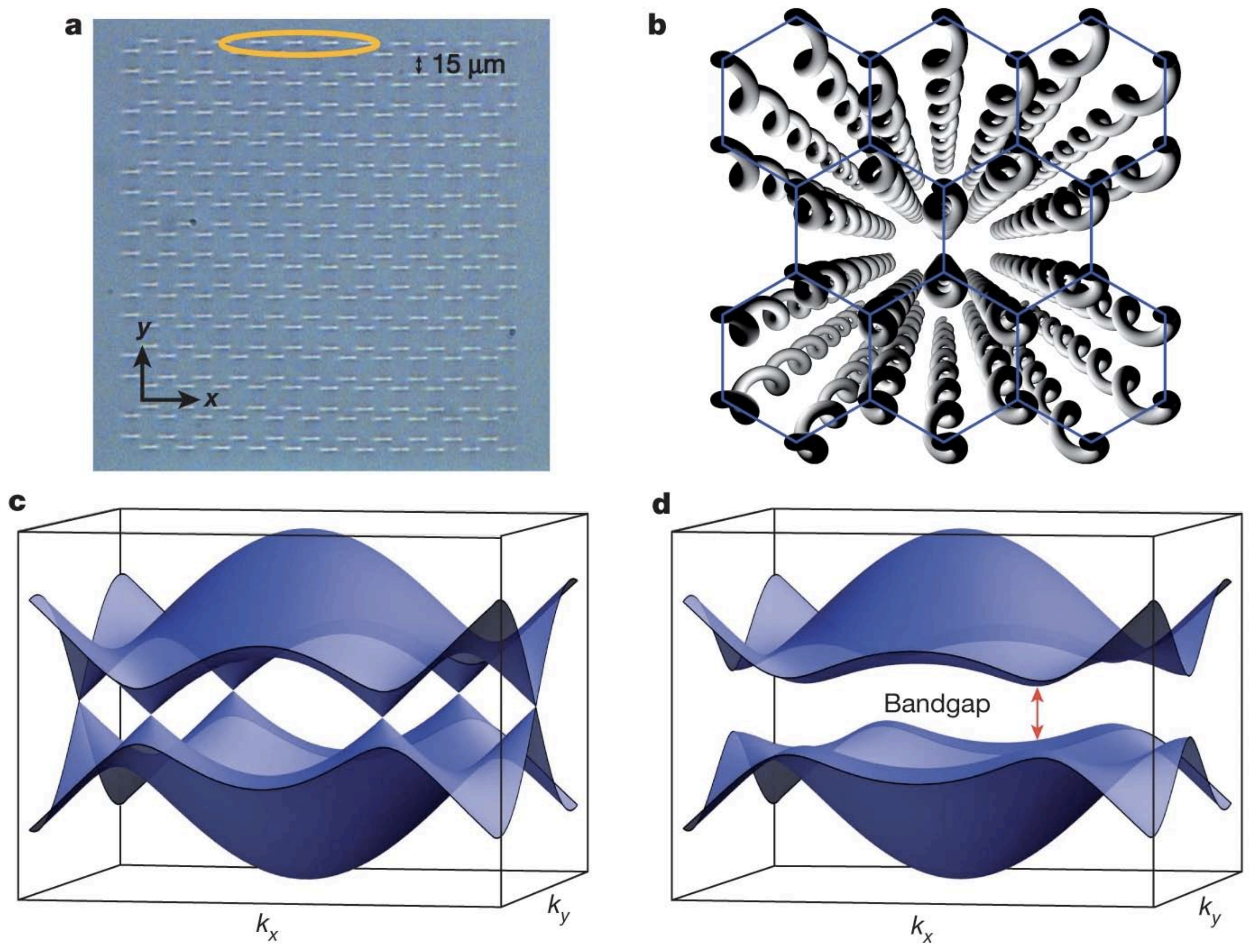
Oka and Aoki, PRB 79, 081406 (2009)

Haldane model emulated with time-periodic drive

Floquet topology across platforms

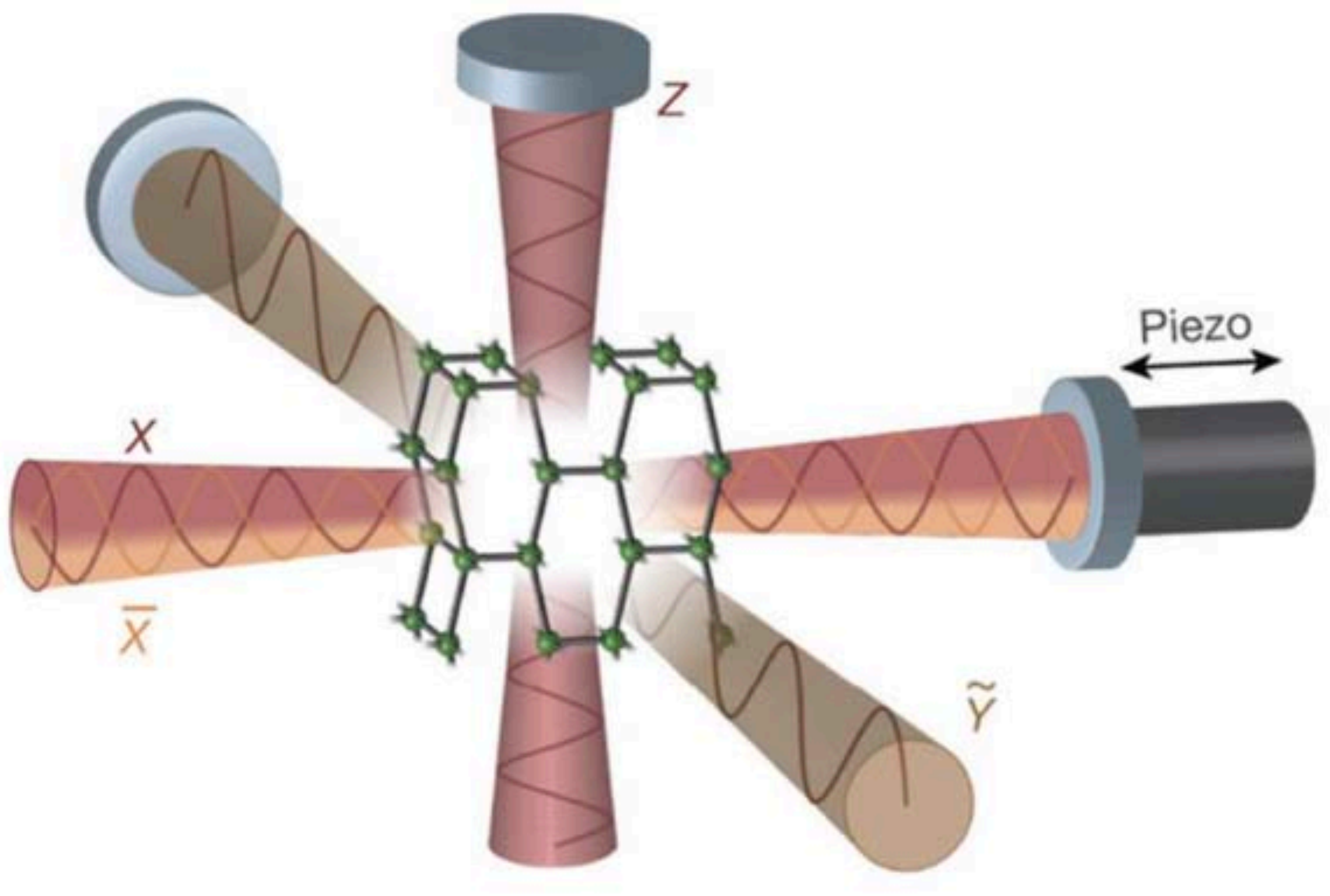
Photonic Floquet topological insulators

M. Rechtsman et al., Nature 496, 196 (2013)



Haldane model with ultracold fermions

G. Jotzu et al., Nature 515, 237 (2014)



review:

M. Rudner and N. Lindner, Nat. Rev. Phys. 2, 229 (2020)

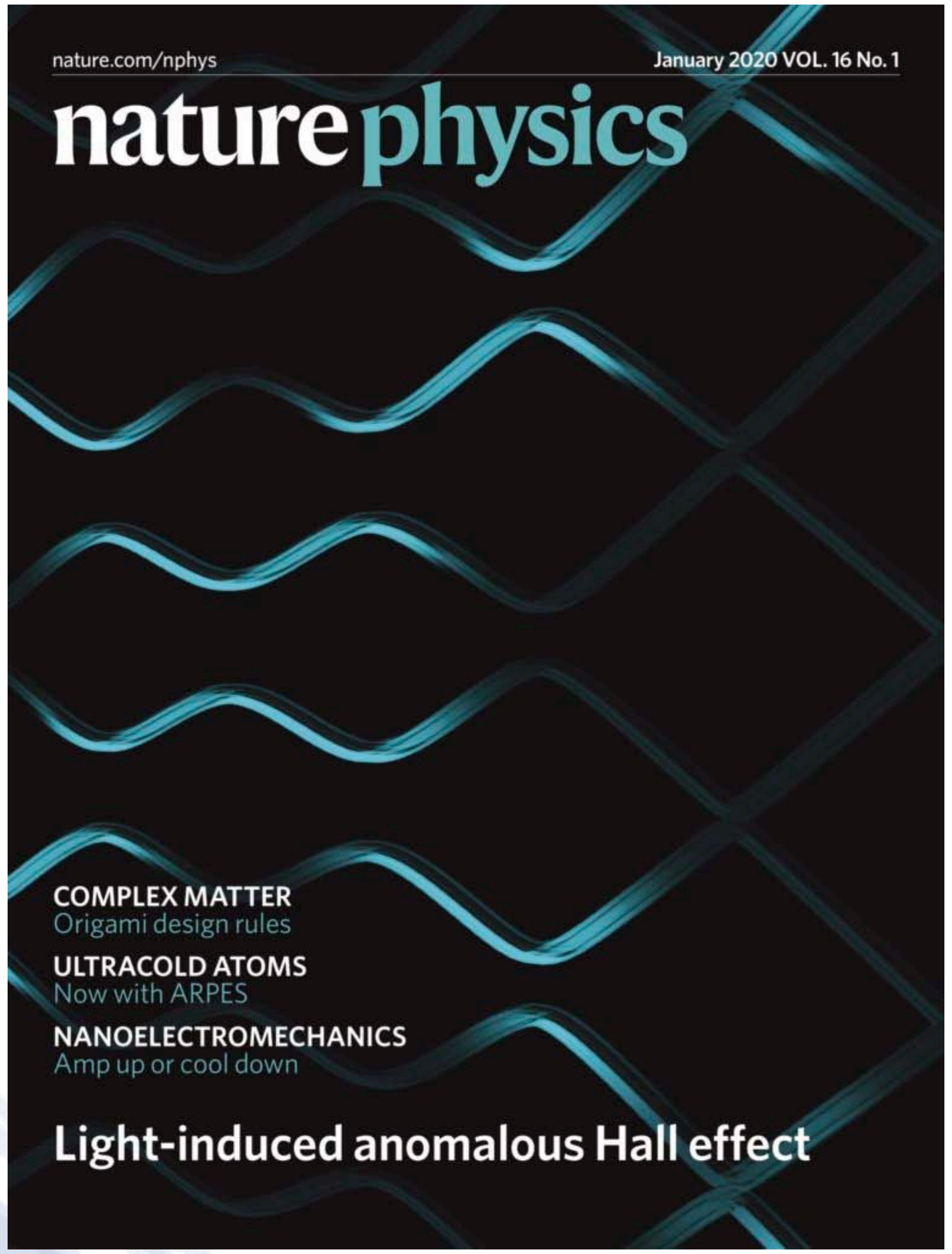
Outline

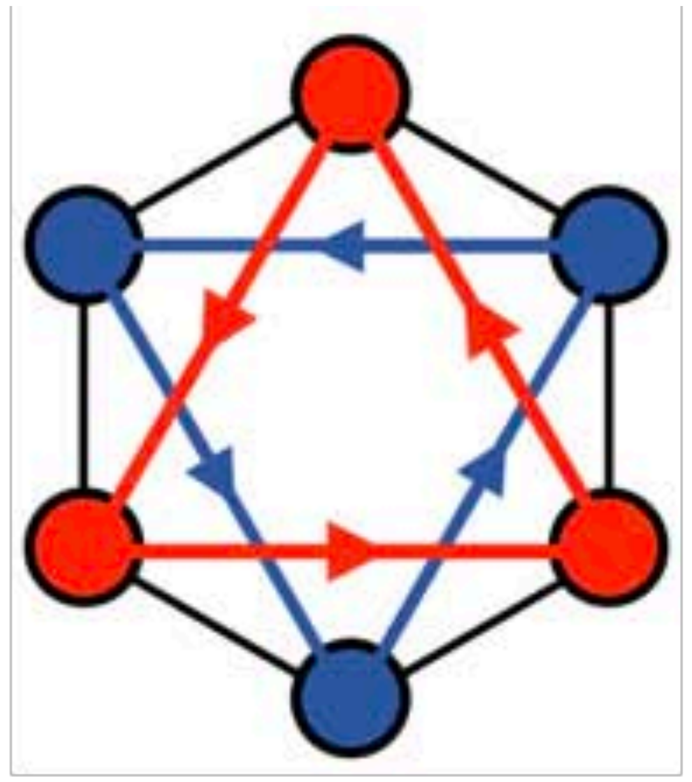
Floquet basics

→ Floquet in solids

Floquet to cavity

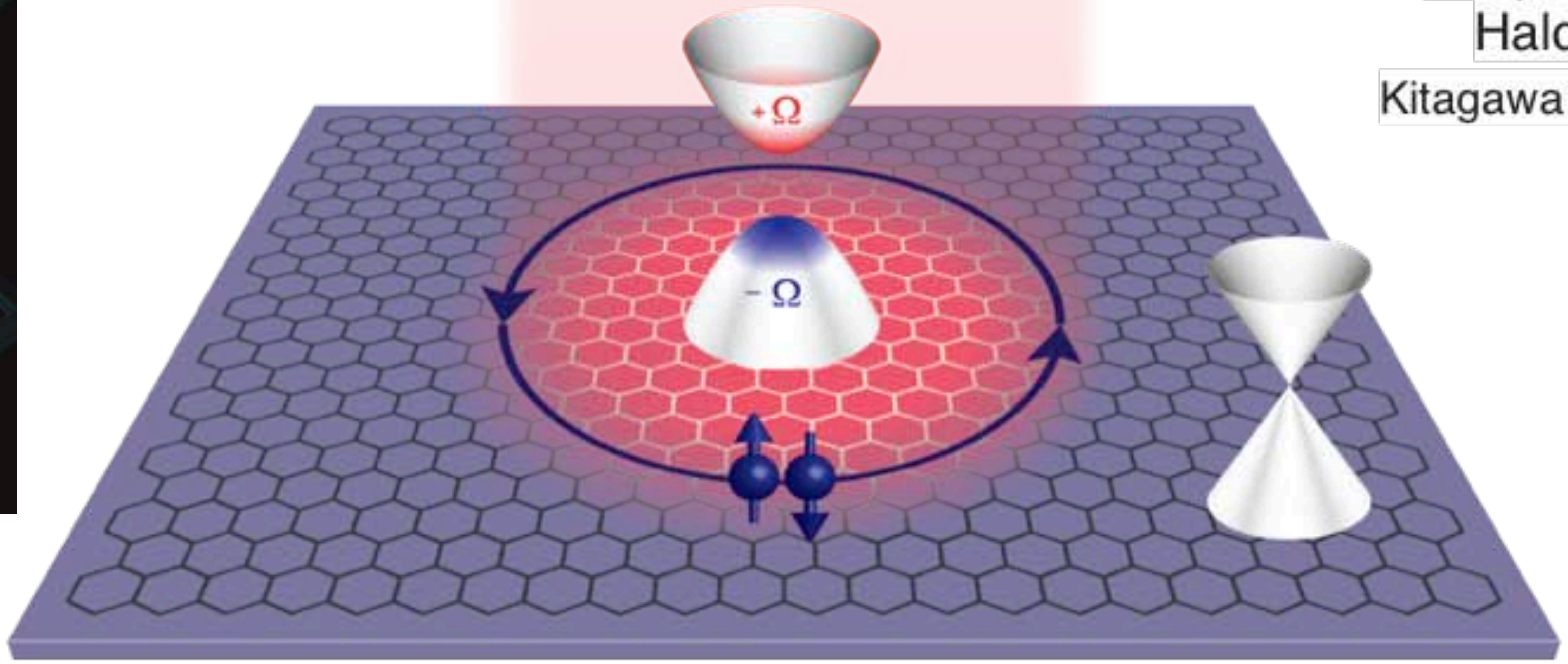
Light-induced anomalous Hall effect





Floquet-engineered Haldane Model

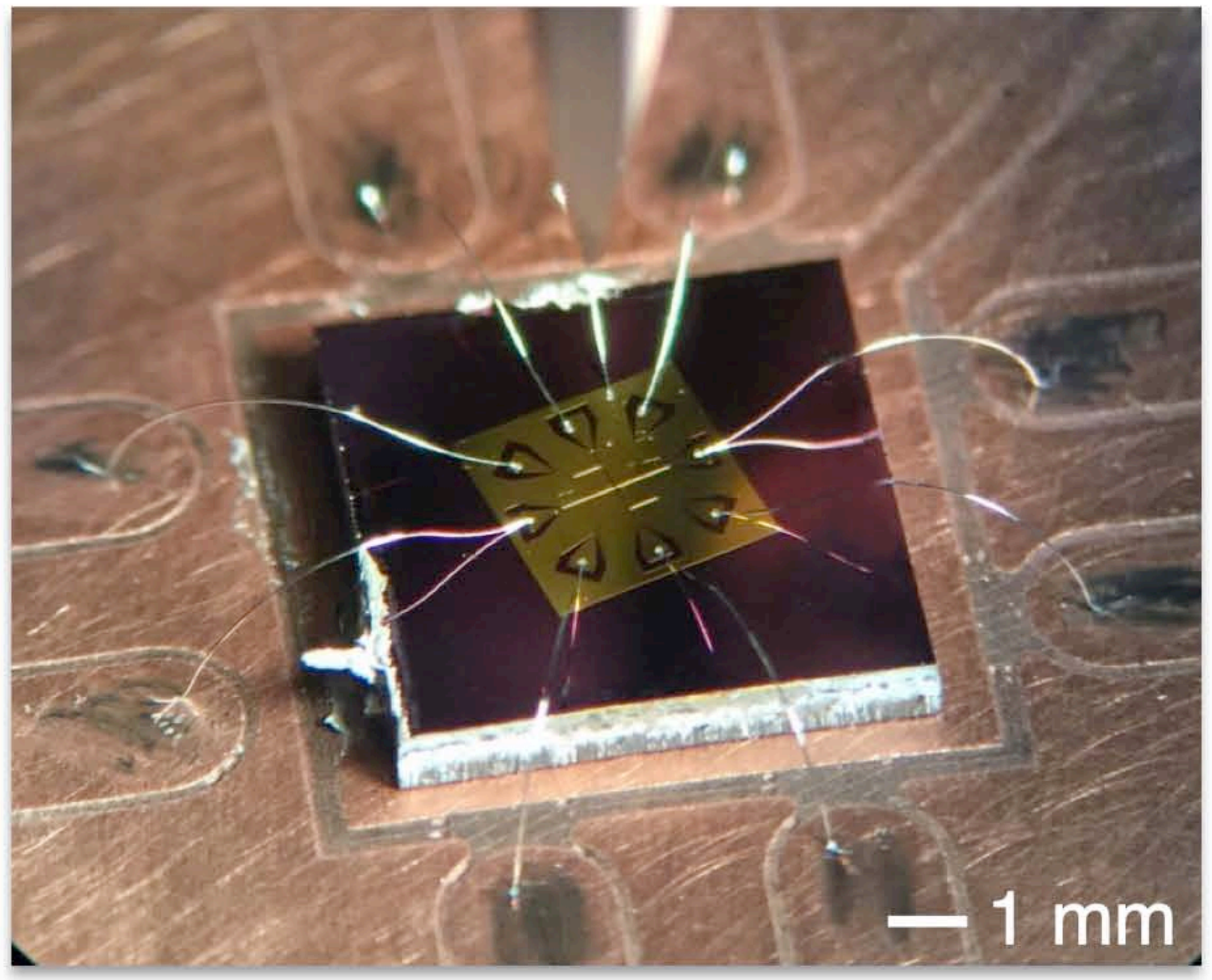
Kitagawa *et al.* PRB (2011)

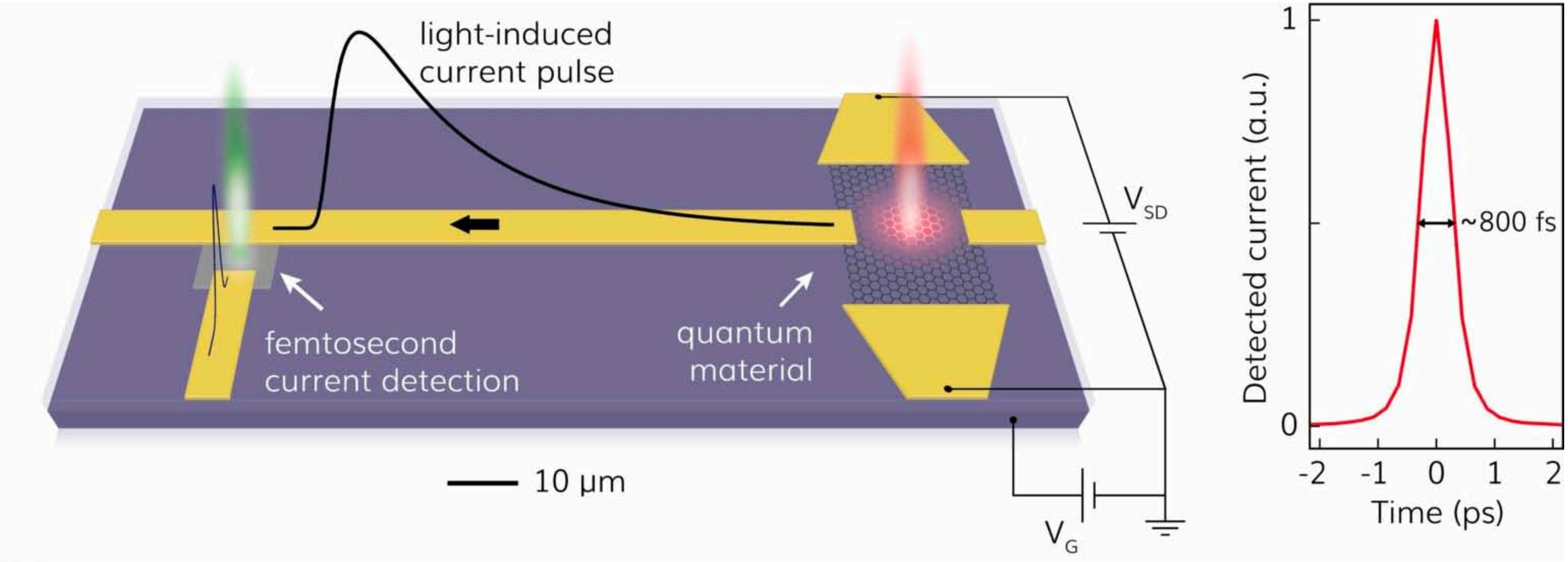


Graphene

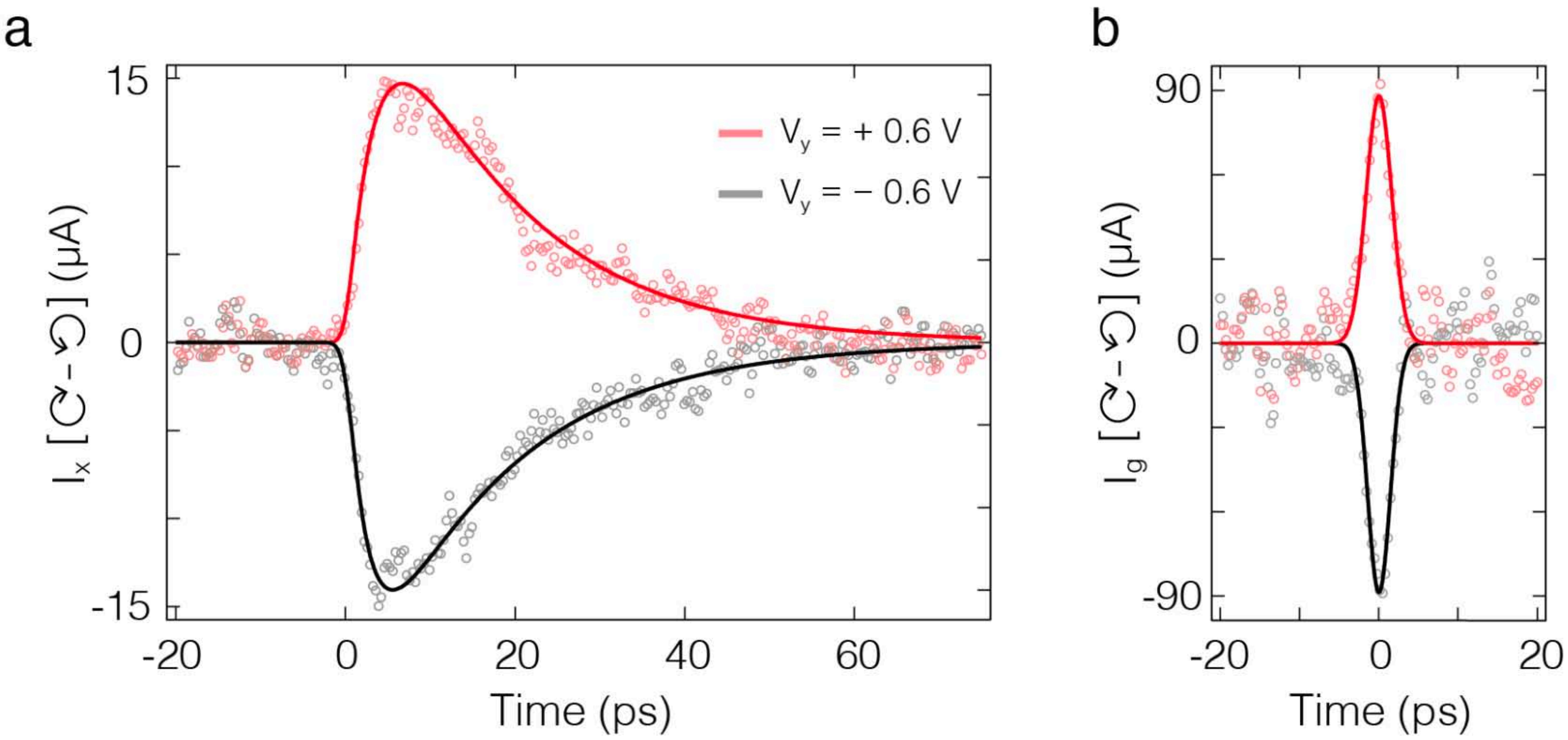
T. Oka & H. Aoki, PRB (2009)

Femtosecond science on-chip

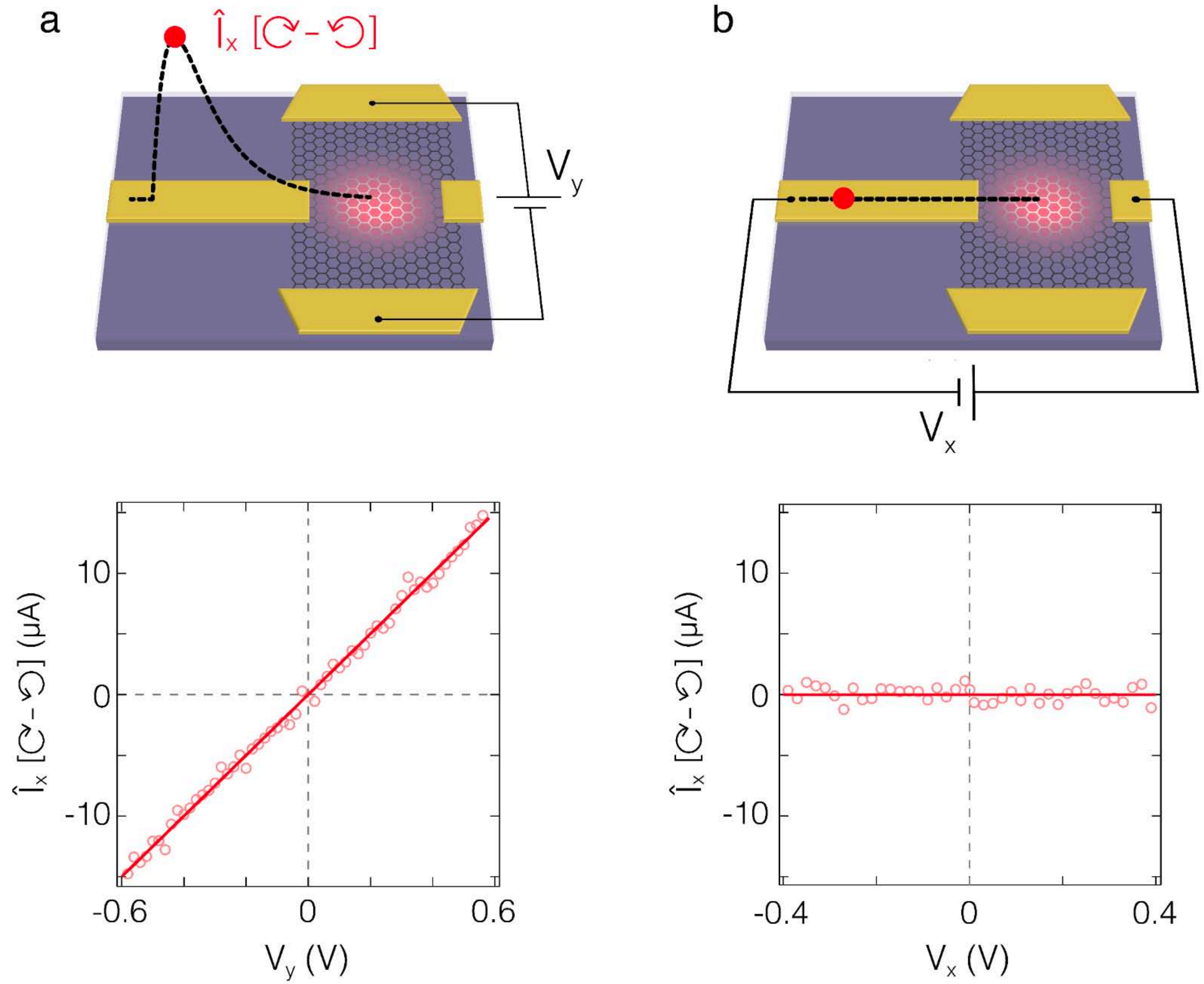




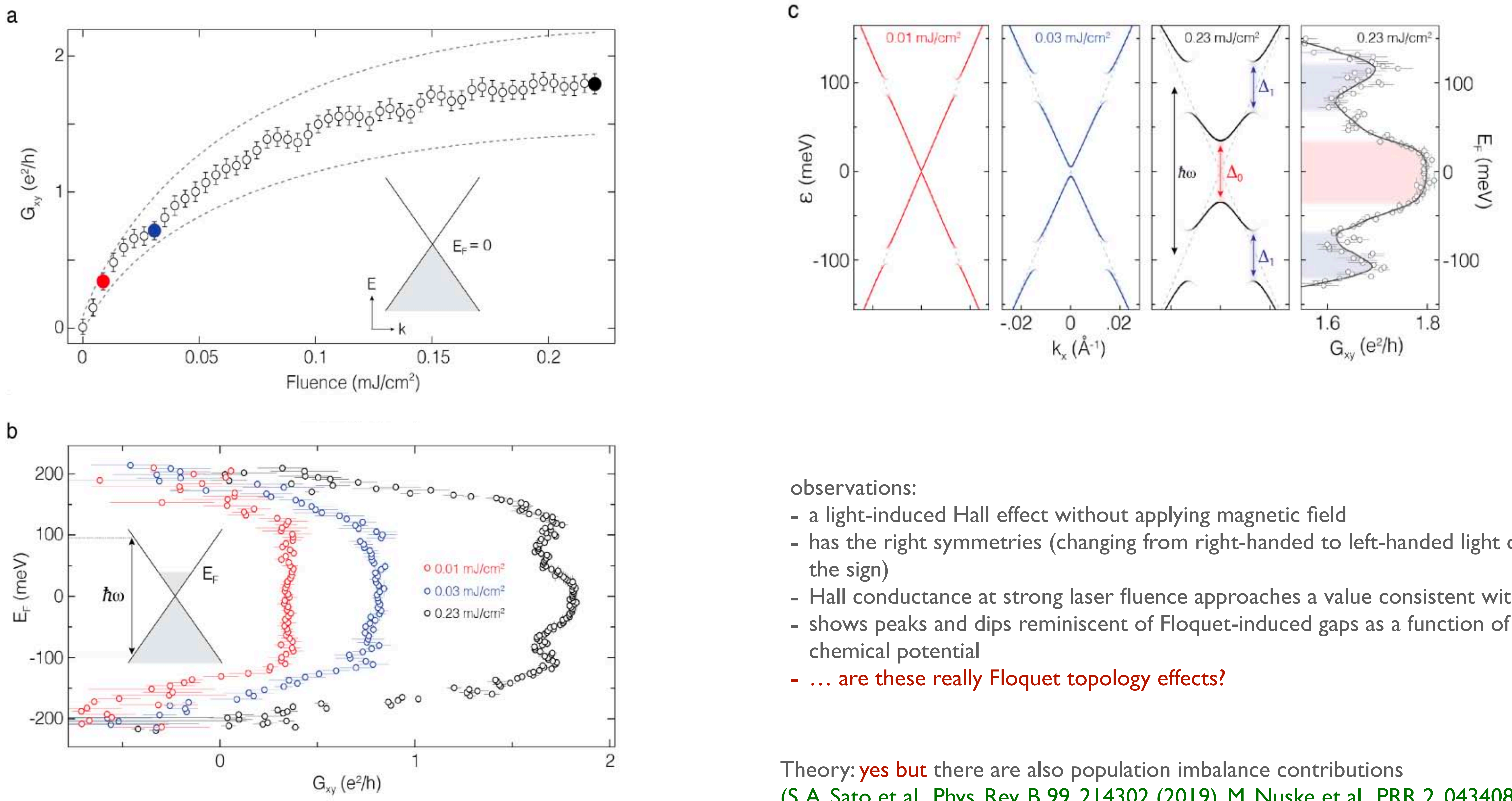
Light-induced anomalous Hall effect



Light-induced anomalous Hall effect



Light-induced anomalous Hall effect



observations:

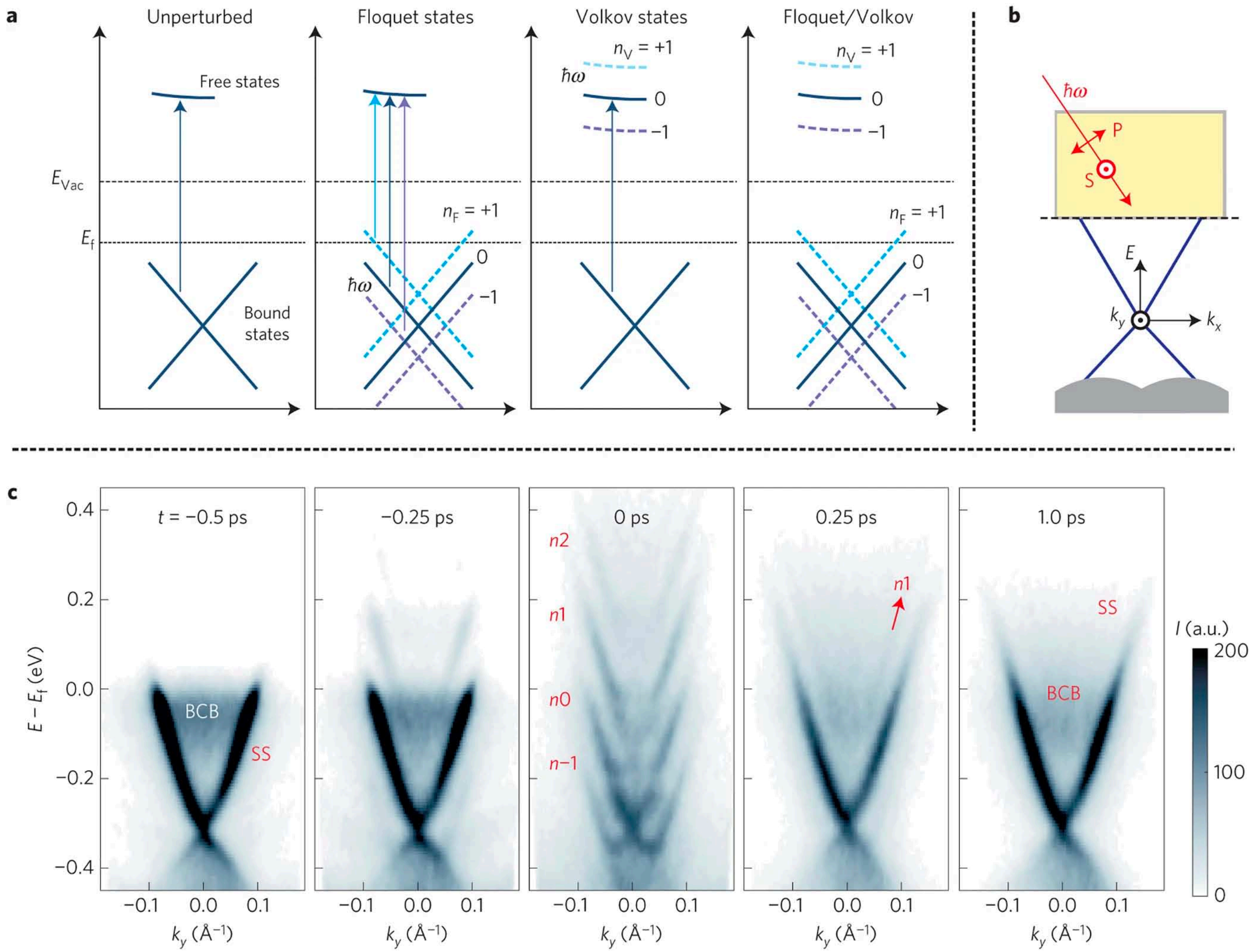
- a light-induced Hall effect without applying magnetic field
- has the right symmetries (changing from right-handed to left-handed light changes the sign)
- Hall conductance at strong laser fluence approaches a value consistent with 2e^2/h
- shows peaks and dips reminiscent of Floquet-induced gaps as a function of chemical potential
- … are these really Floquet topology effects?

Theory: yes but there are also population imbalance contributions
(S.A. Sato et al., Phys. Rev. B 99, 214302 (2019), M. Nuske et al., PRR 2, 043408 (2020))

Y. H. Wang et al., Science 342, 453 (2013)
F. Mahmood et al., Nature Physics 12, 306-310 (2016)
[Nuh Gedik group at MIT]

Floquet physics in materials

… observation in 3D topological insulator surface 2D Dirac fermions

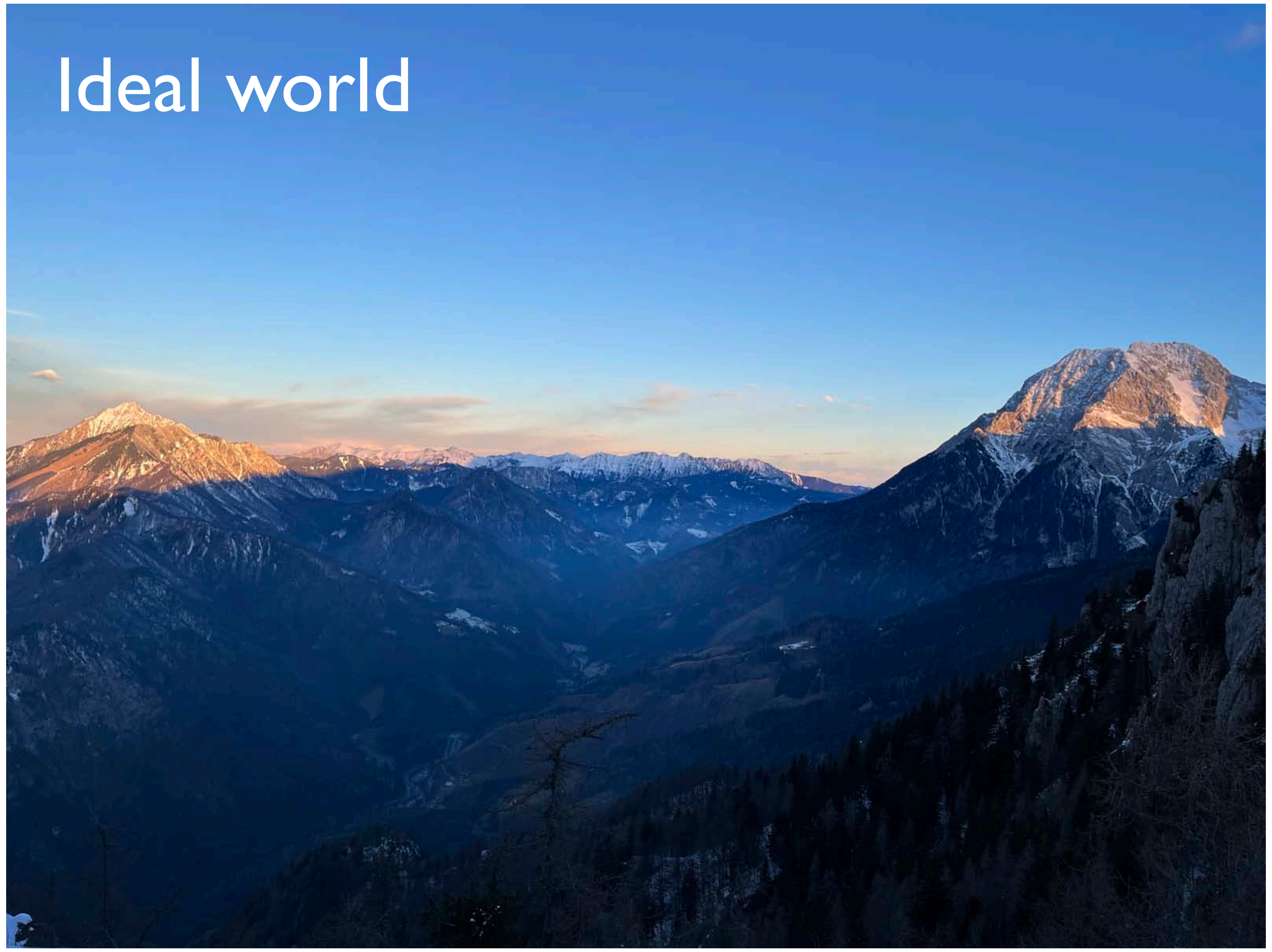


What about observing Floquet states in time-resolved photoemission of graphene?

Ongoing challenge to overcome decoherence and dissipation

e.g., Aeschlimann et al., Nano Lett. 21, 5028 (2021)

… but stay tuned!



Floquet engineering as flexible scheme to dynamically create „new quantum materials“

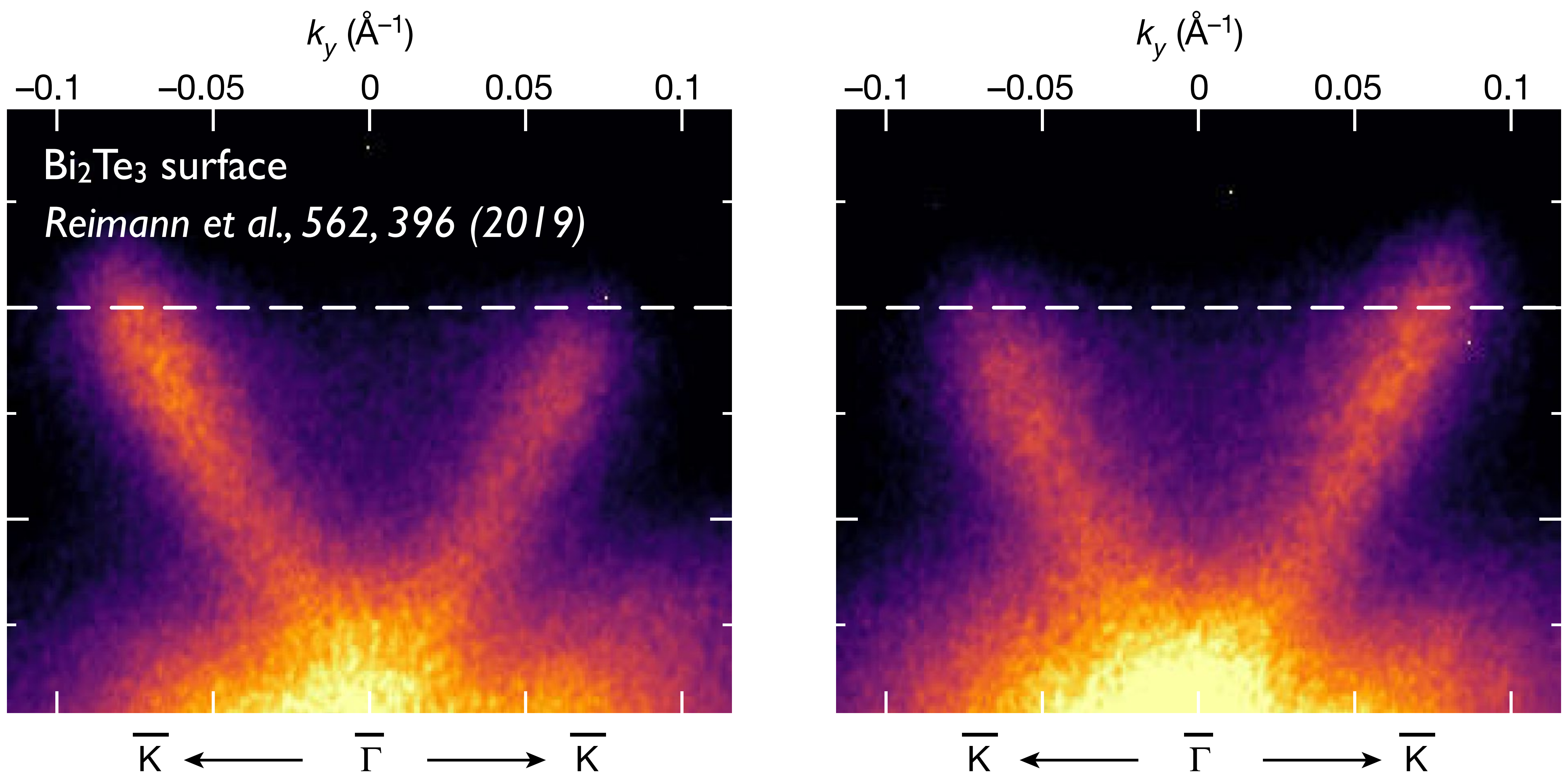
Floquet effects very hard to observe

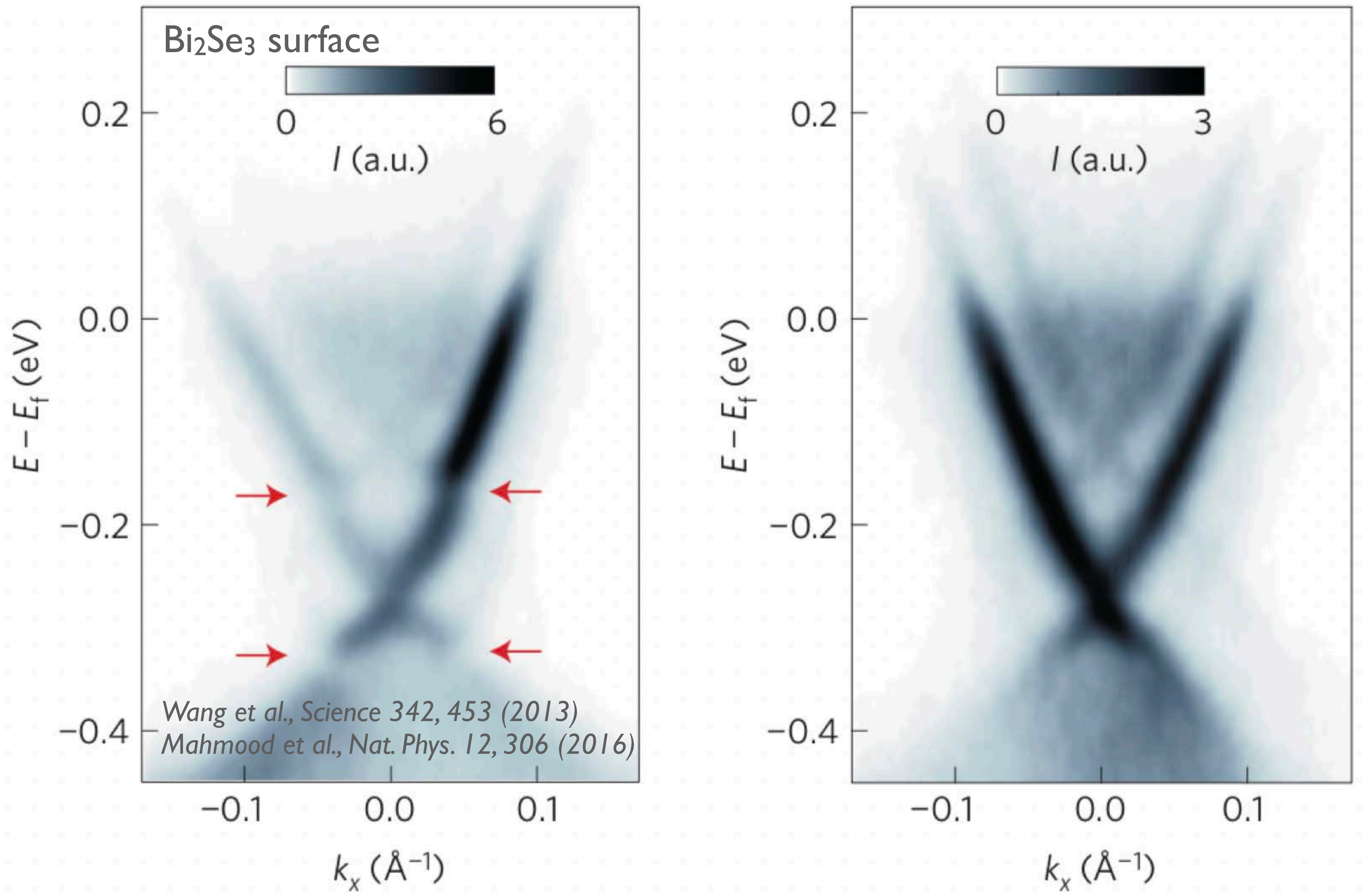
heating, decoherence, …

(A) use strong fields but probe faster: subcycle

(B) use strong coupling instead of strong fields (hence, cavities!)

A fundamental question in strong fields





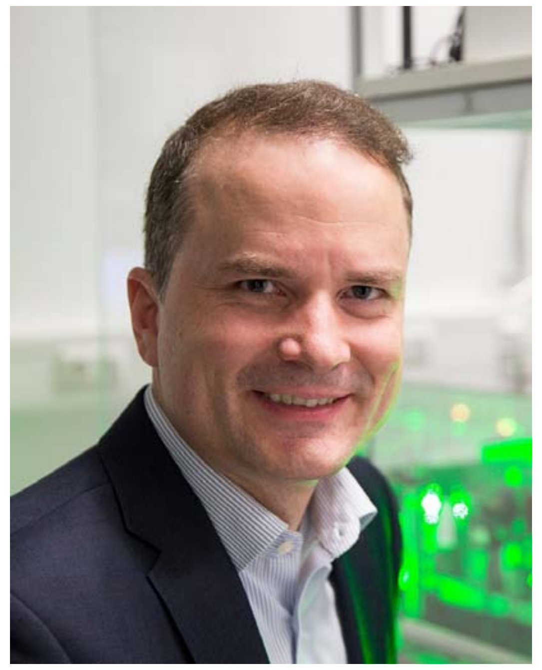
?

subcycle ⟷ many cycle (Floquet)

Buildup and dephasing of Floquet-Bloch bands on subcycle time scales

S. Ito[1,†], M. Schüler[2,†], M. Meierhofer[3], S. Schlauderer[3], J. Freudenstein[3], J. Reimann[1], D. Afanasiev[3], K. A. Kokh[5], O. E. Tereshchenko[5], J. Güdde[1], M. A. Sentef[4,*], U. Höfer[1,3,*], R. Huber[3,*]

Nature 616, 696–701 (2023)

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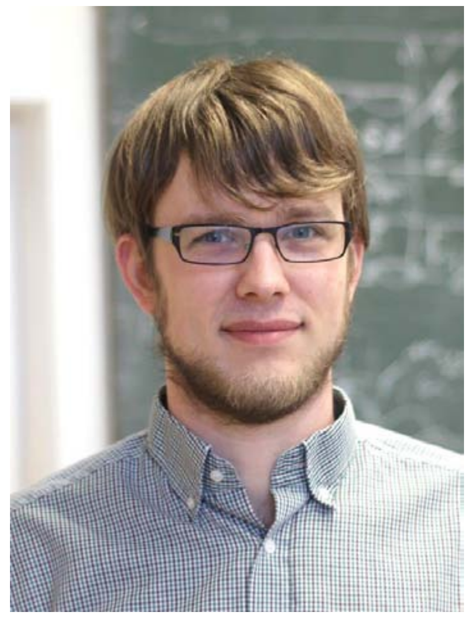
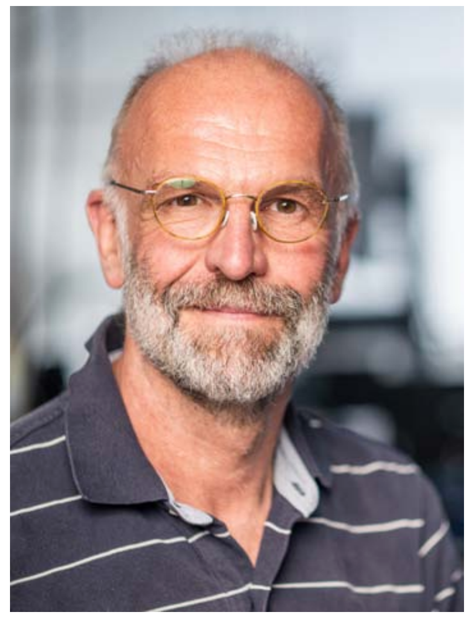
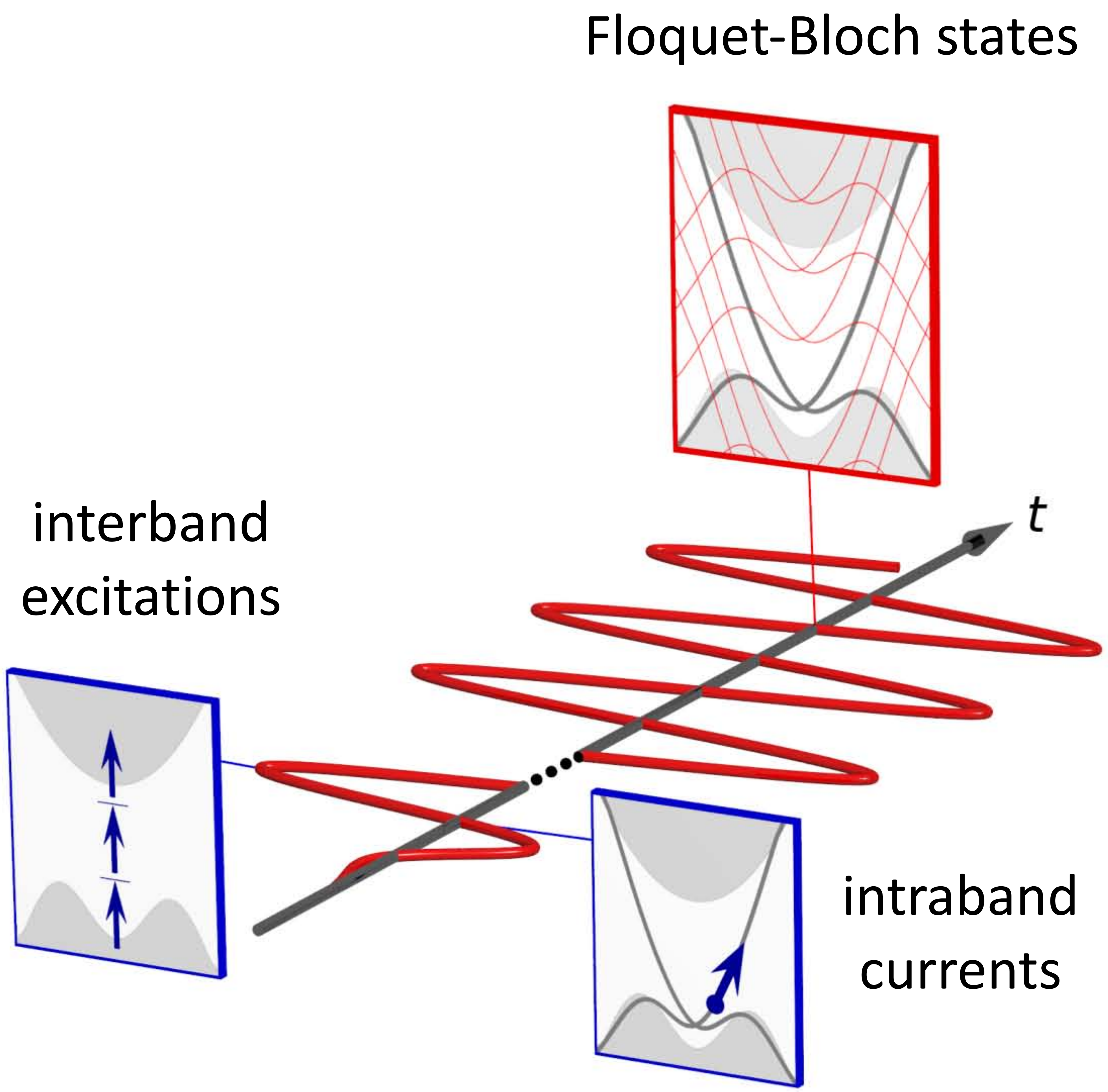


Strong-field phenomena in a unified picture

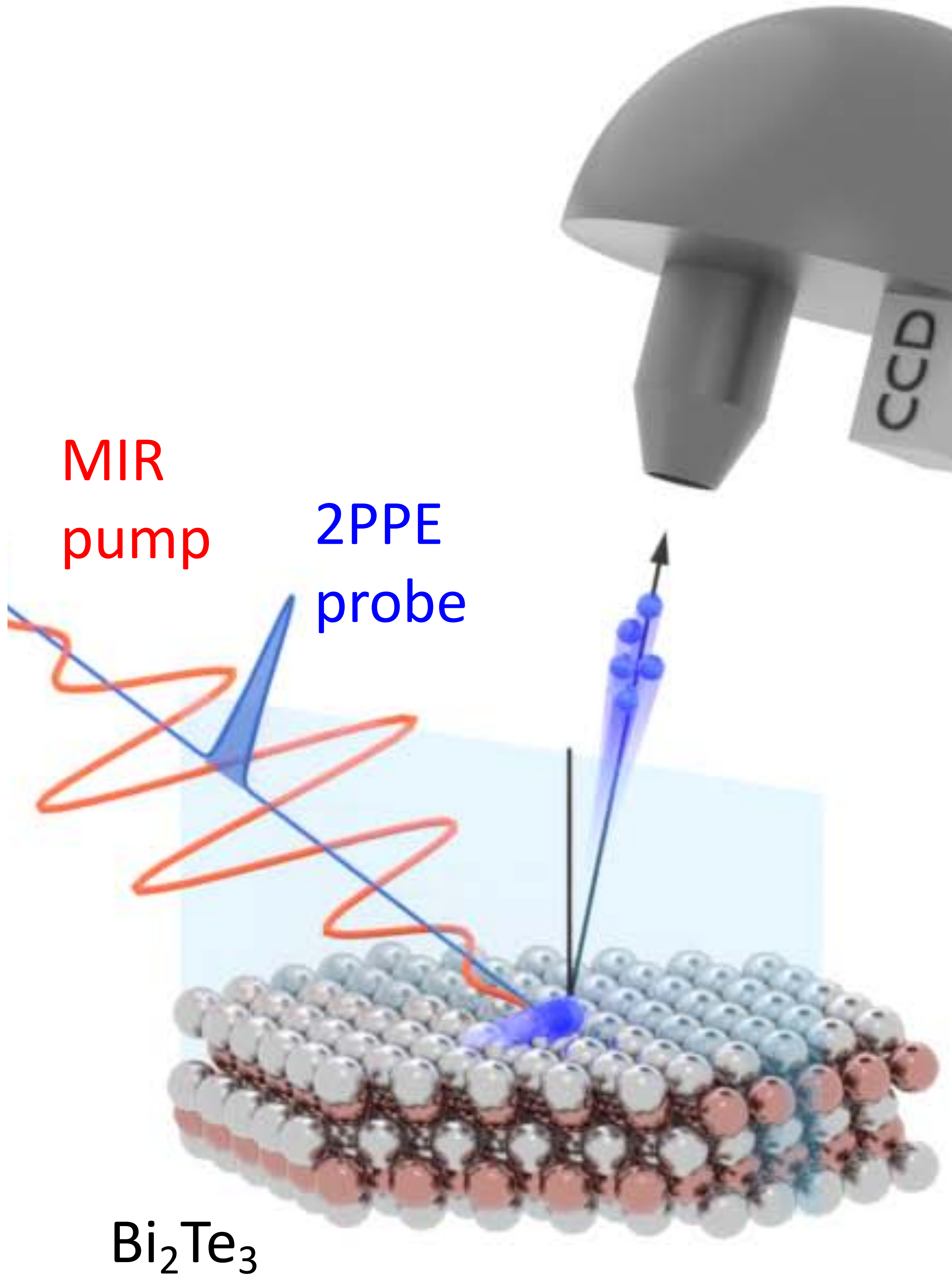


Questions

- how are these notions connected?
- how and when do Floquet states emerge?

Toward the strong-field regime

Extending subcycle ARPES to the MV/cm range



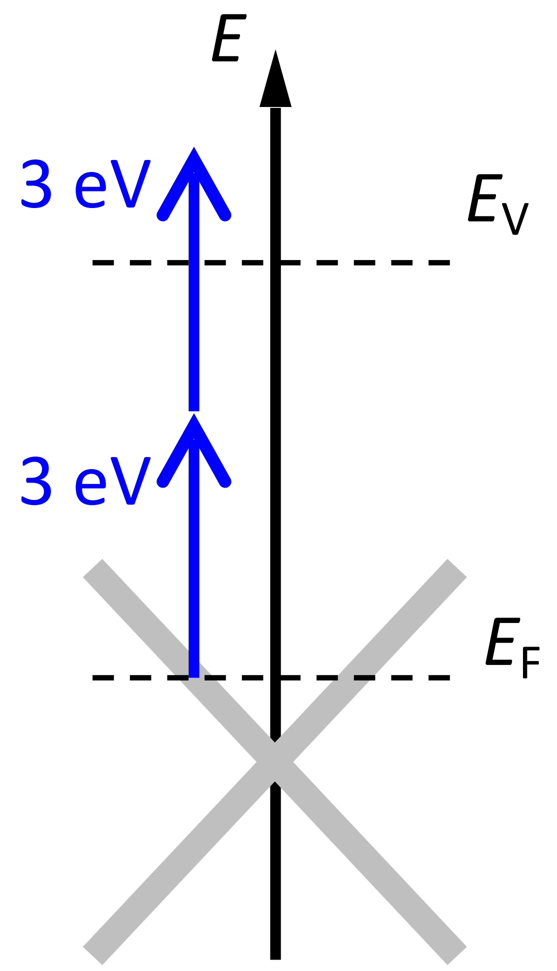
Use of mid-infrared (MIR) pump: 20-40 THz

- can reach MV/cm field strength

  [Sell, Leitenstorfer, Huber, Opt. Lett. **33**, 2767 (2008)]

- 1 optical cycle: 25-40 fs

Use of two-photon photoemission (2PPE) probe



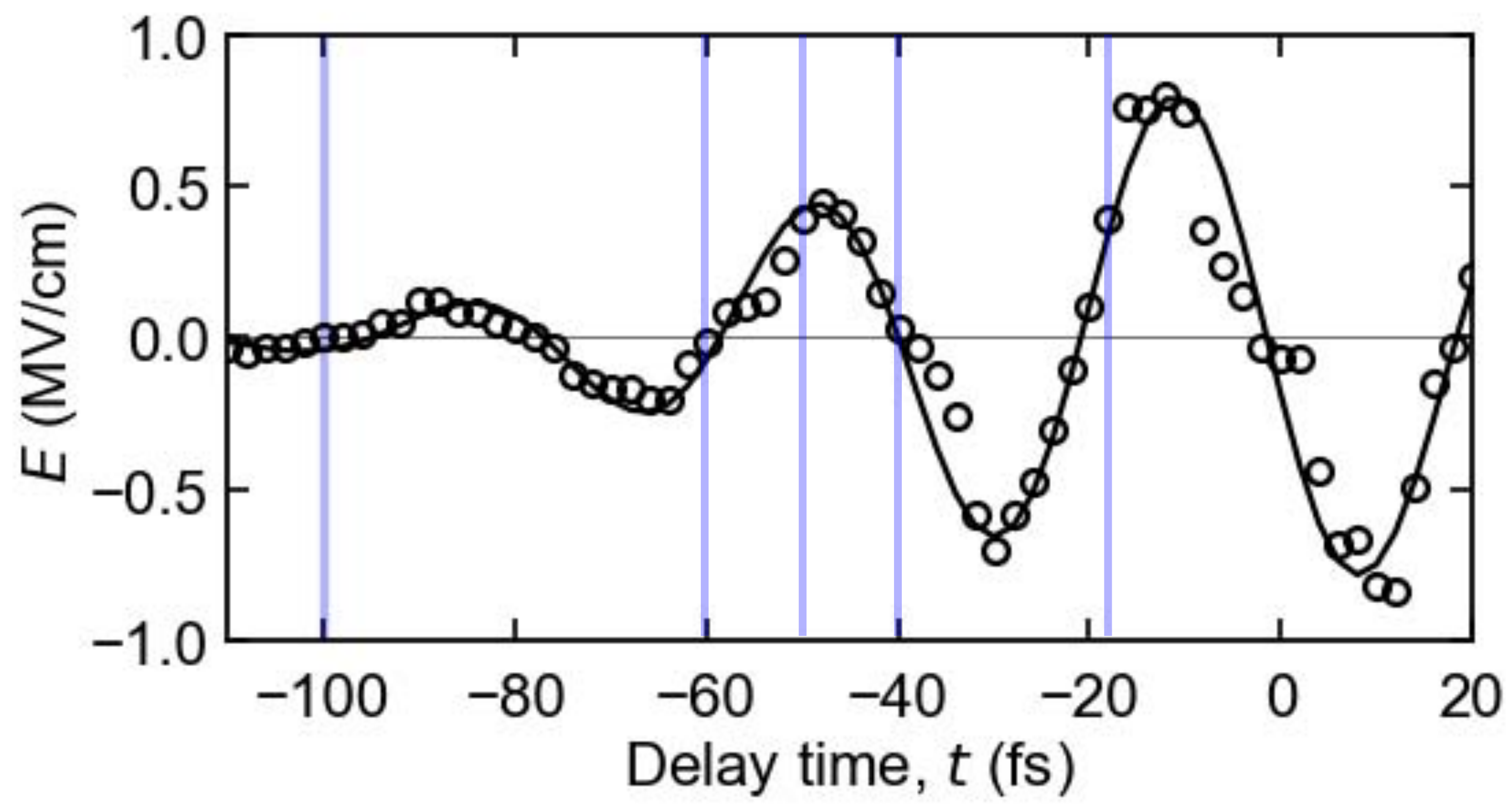
10-20 fs, 3 eV pulse is feasible

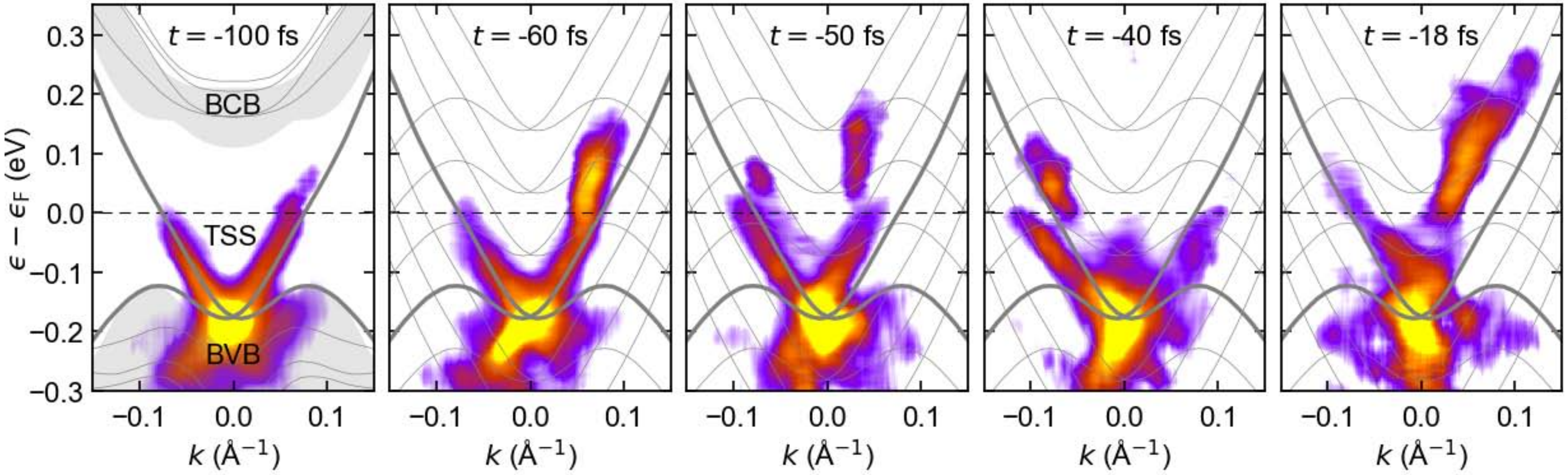
Subcycle ARPES in the strong-field regime

Lightwave-driven dynamics in momentum space

Streaking-compensated ARPES images

→ **show intrinsic dynamics in matter**





lightwave-driven currents + subcycle band engineering!

“Birth” of Floquet-Bloch states - Theory

Fully quantum Tr-ARPES calculations

Michael Schüler

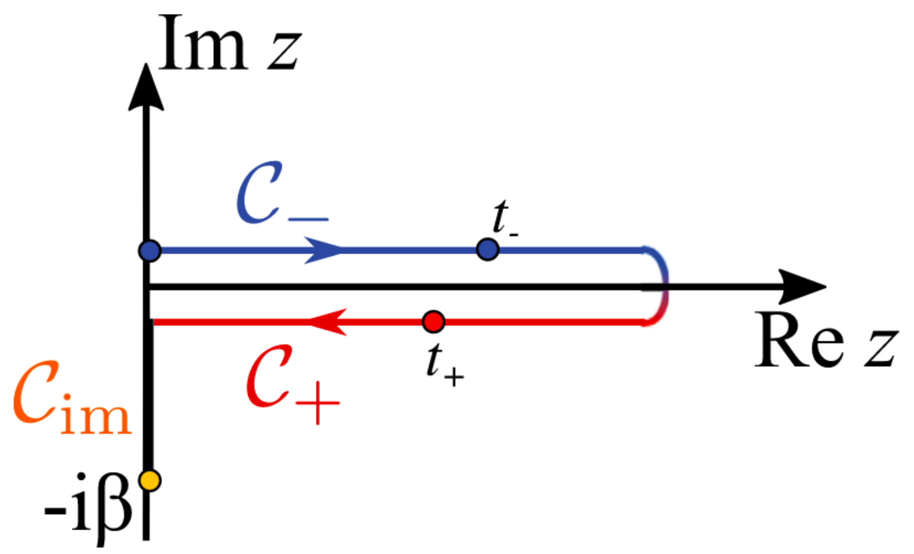
- non-equilibrium Green-function formalism
- use the experimental pump waveform
- include ARPES matrix elements
- phenomenologically consider the 2PPE probe

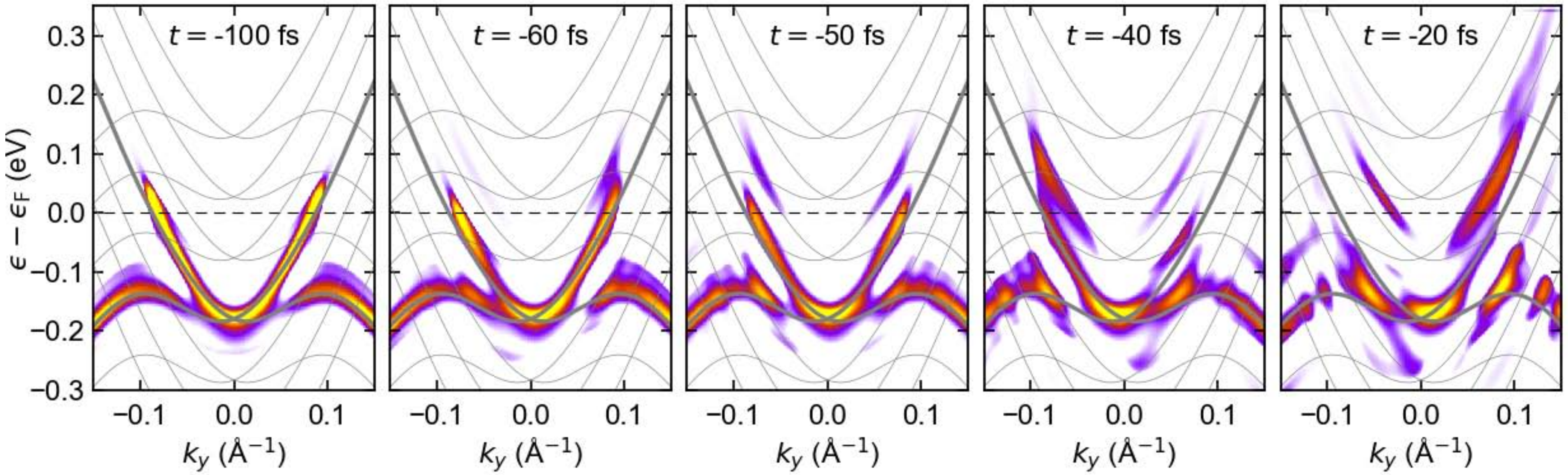
For formalism see

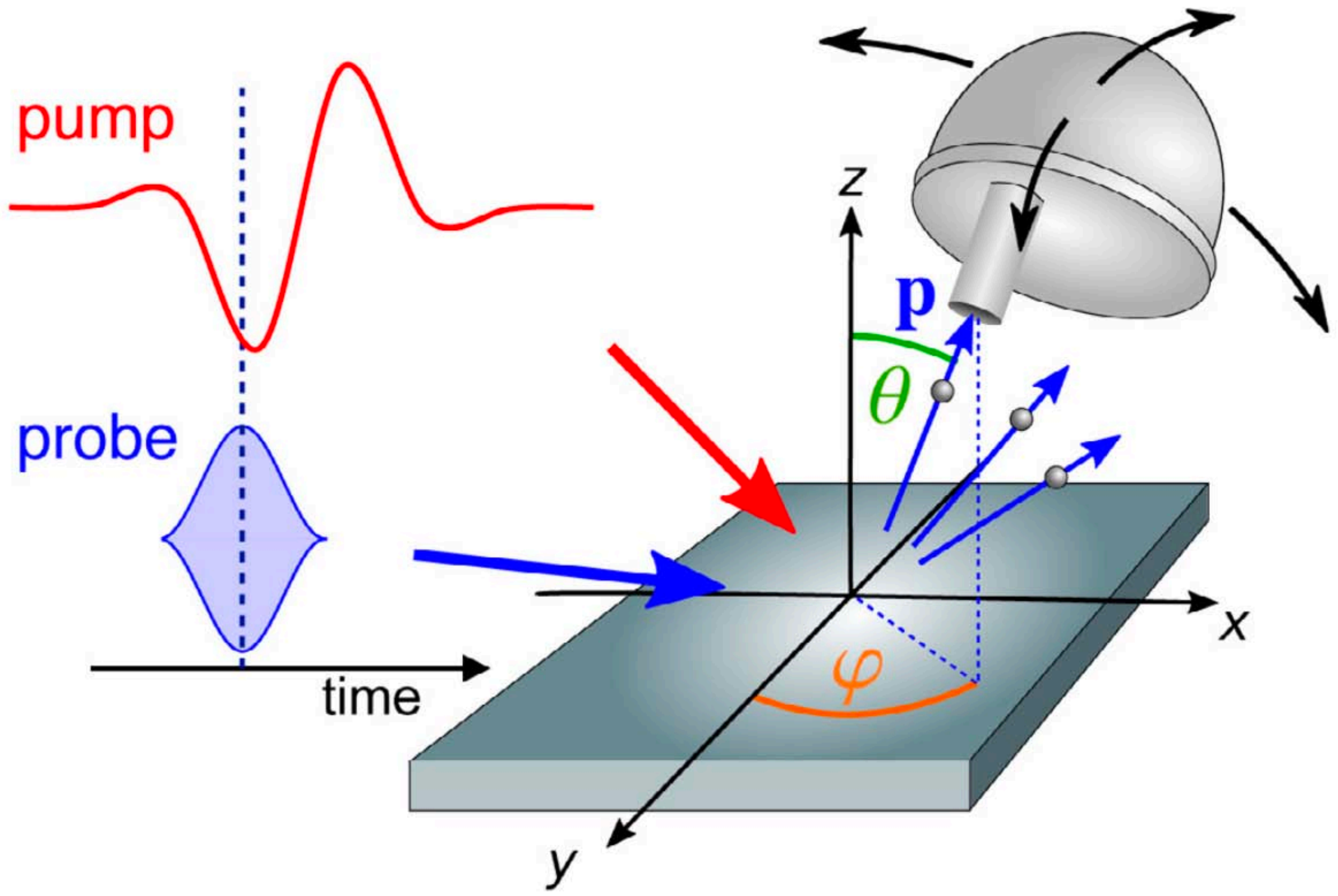
Theory of subcycle time-resolved photoemission: Application to terahertz photodressing in graphene

Michael Schüler [a], Michael A. Sentef [b]

https://doi.org/10.1016/j.elspec.2021.147121

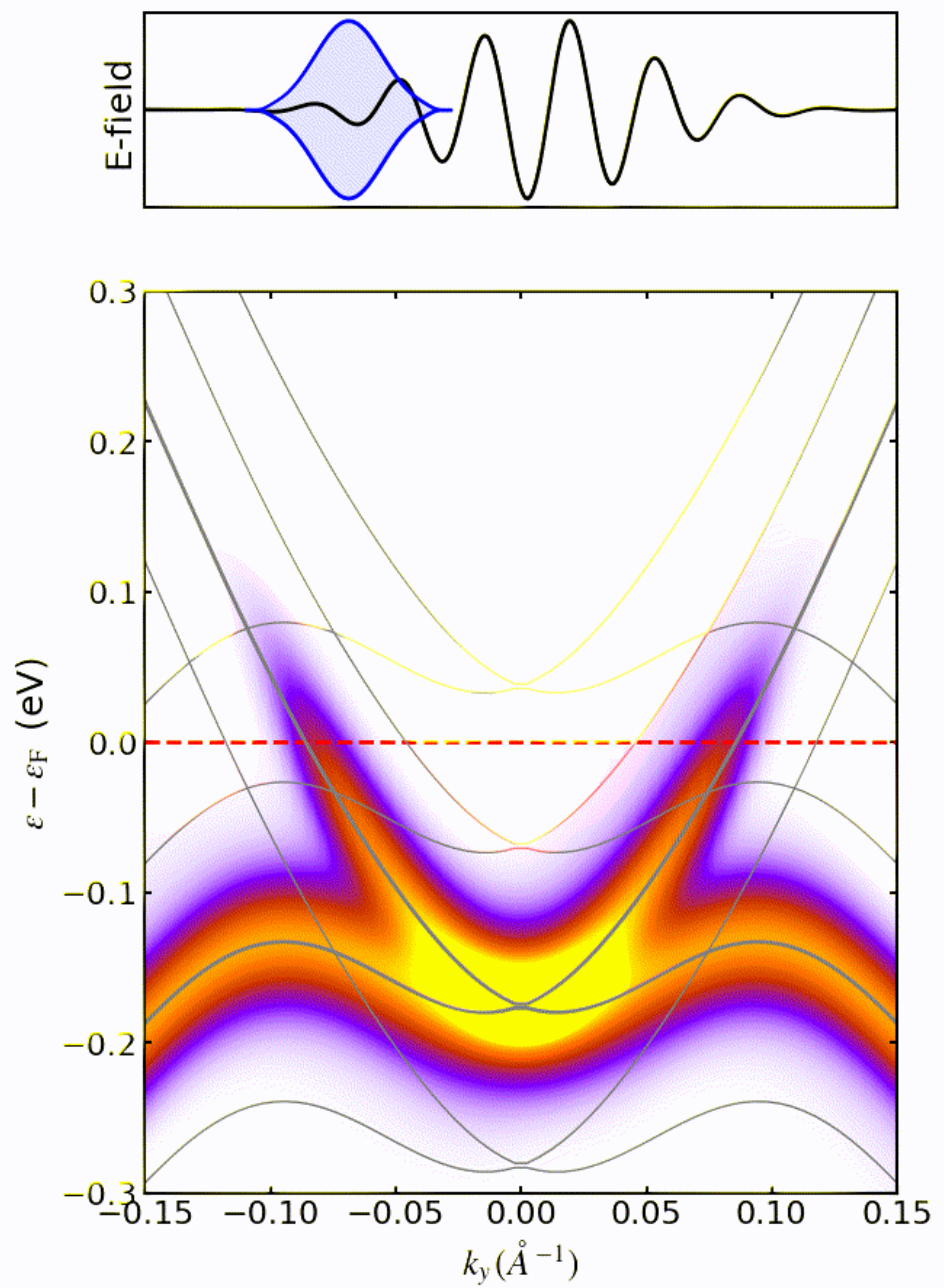




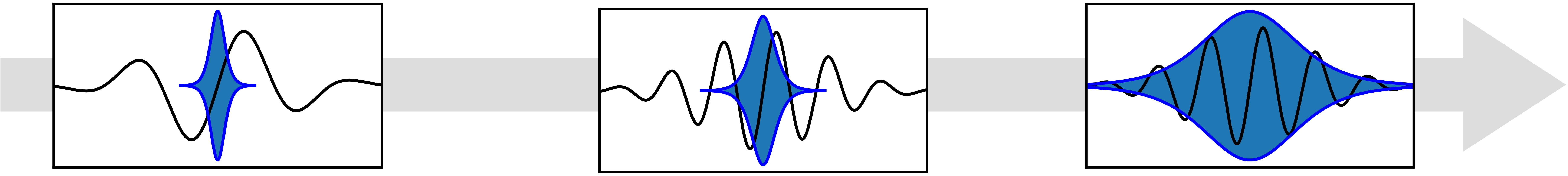


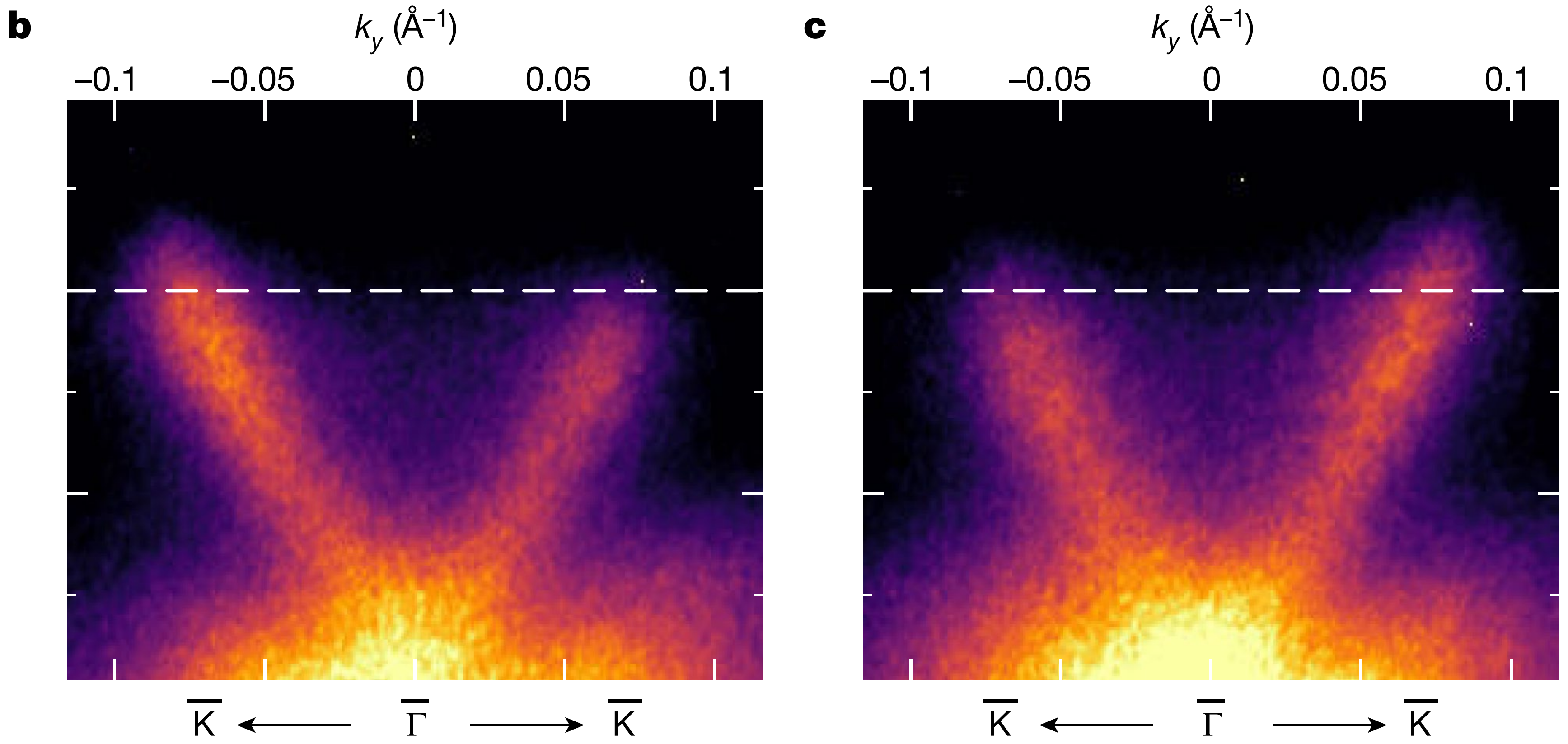
Subcycle evolution of Floquet-Bloch states is explained by theory!

Michael Schüler

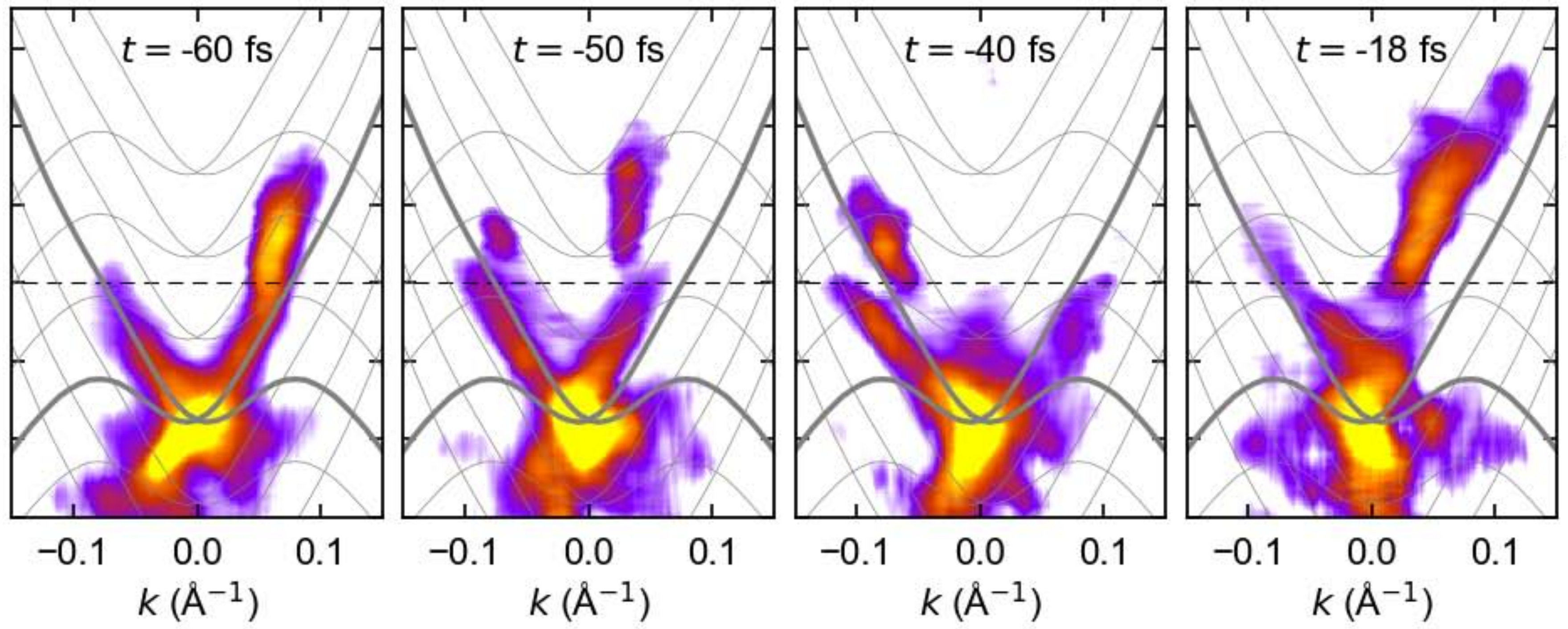


Quasiadiabatic regime — Subcycle regime — Floquet regime

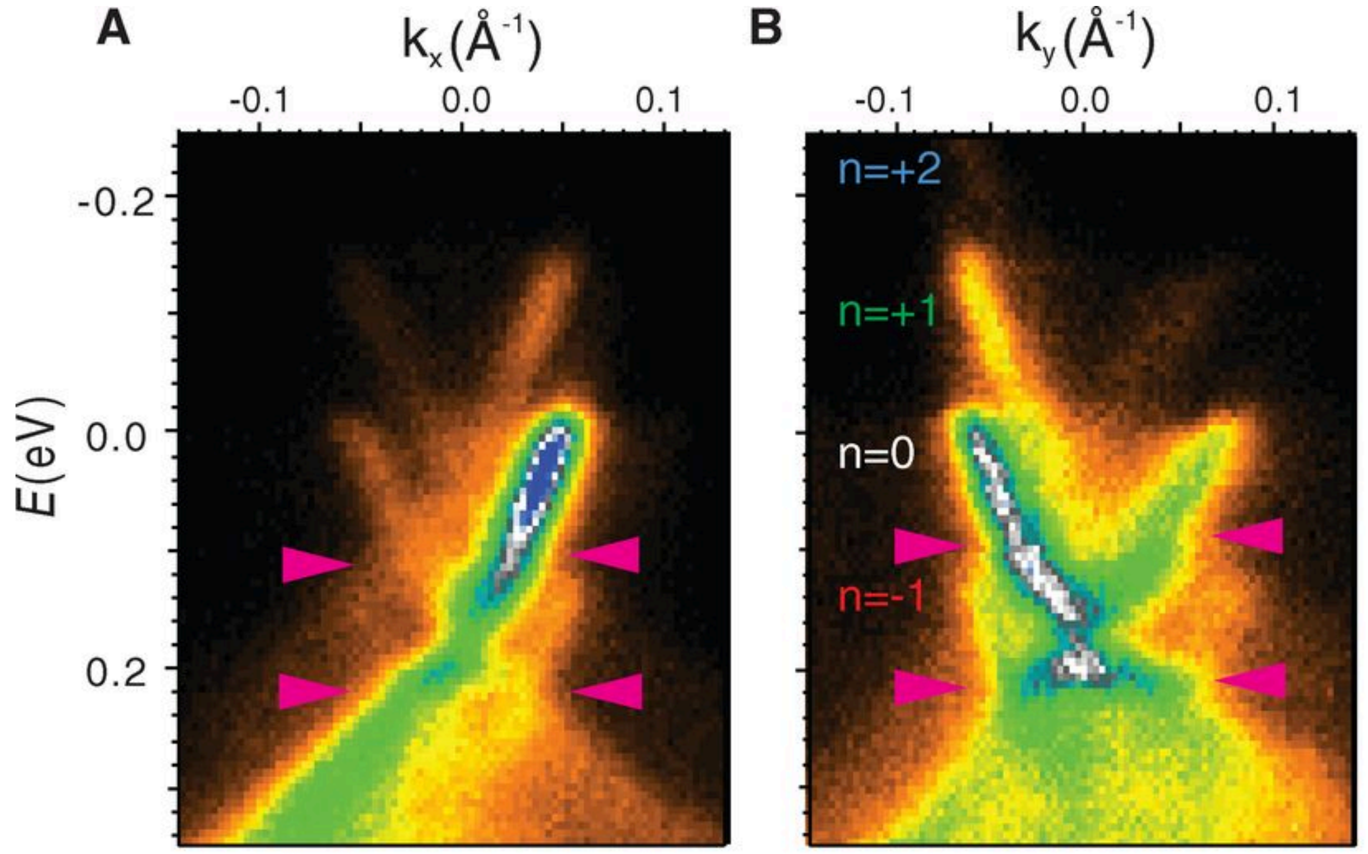




Reimann et al., Nature **562**, 396 (2019)



Ito, Schüler, et al., Nature **616**, 696 (2023)



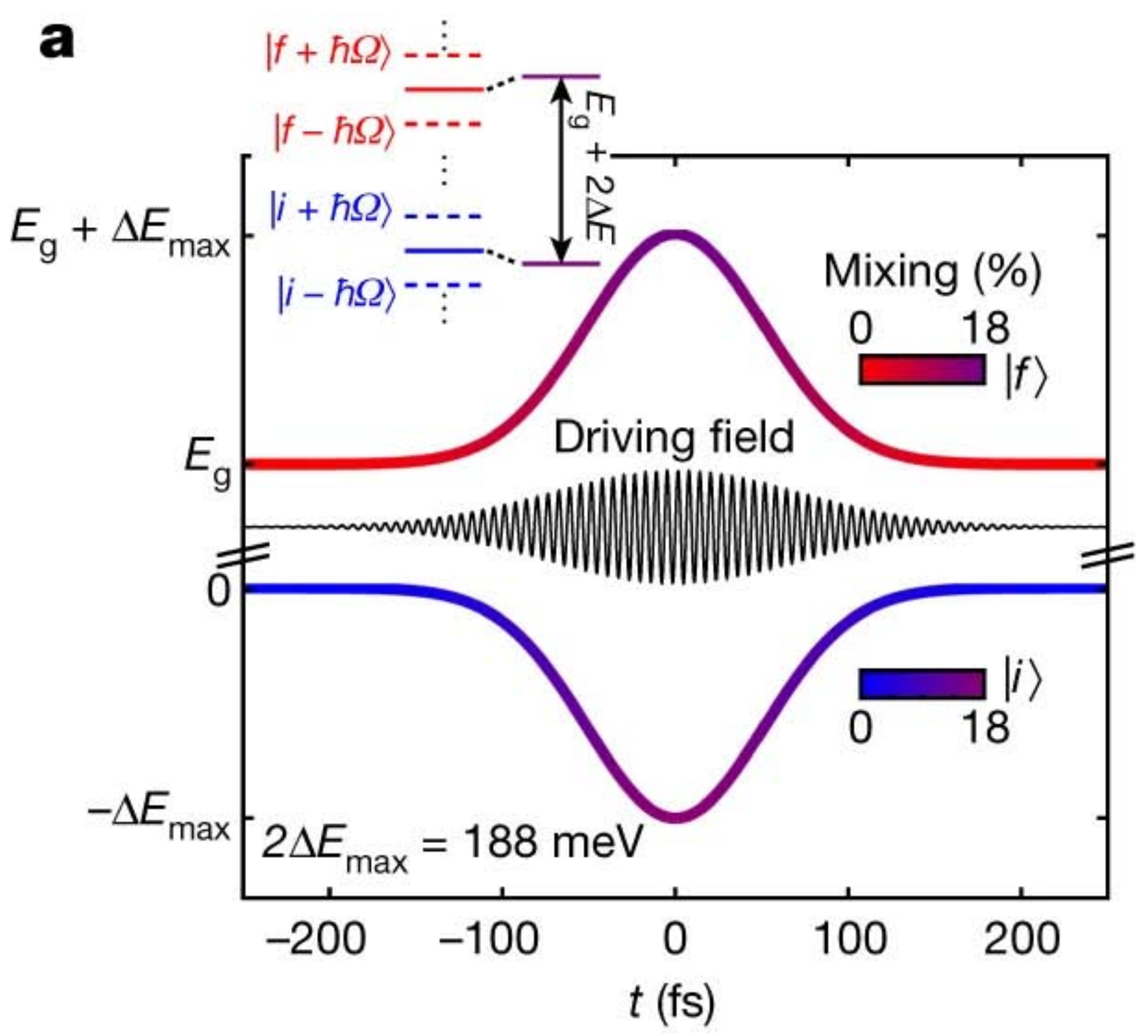
Wang et al., Science **342**, 453 (2013)

Opens new opportunities to dynamically control emergent properties and functionality on subcycle time scales

Floquet materials science

MnPS3

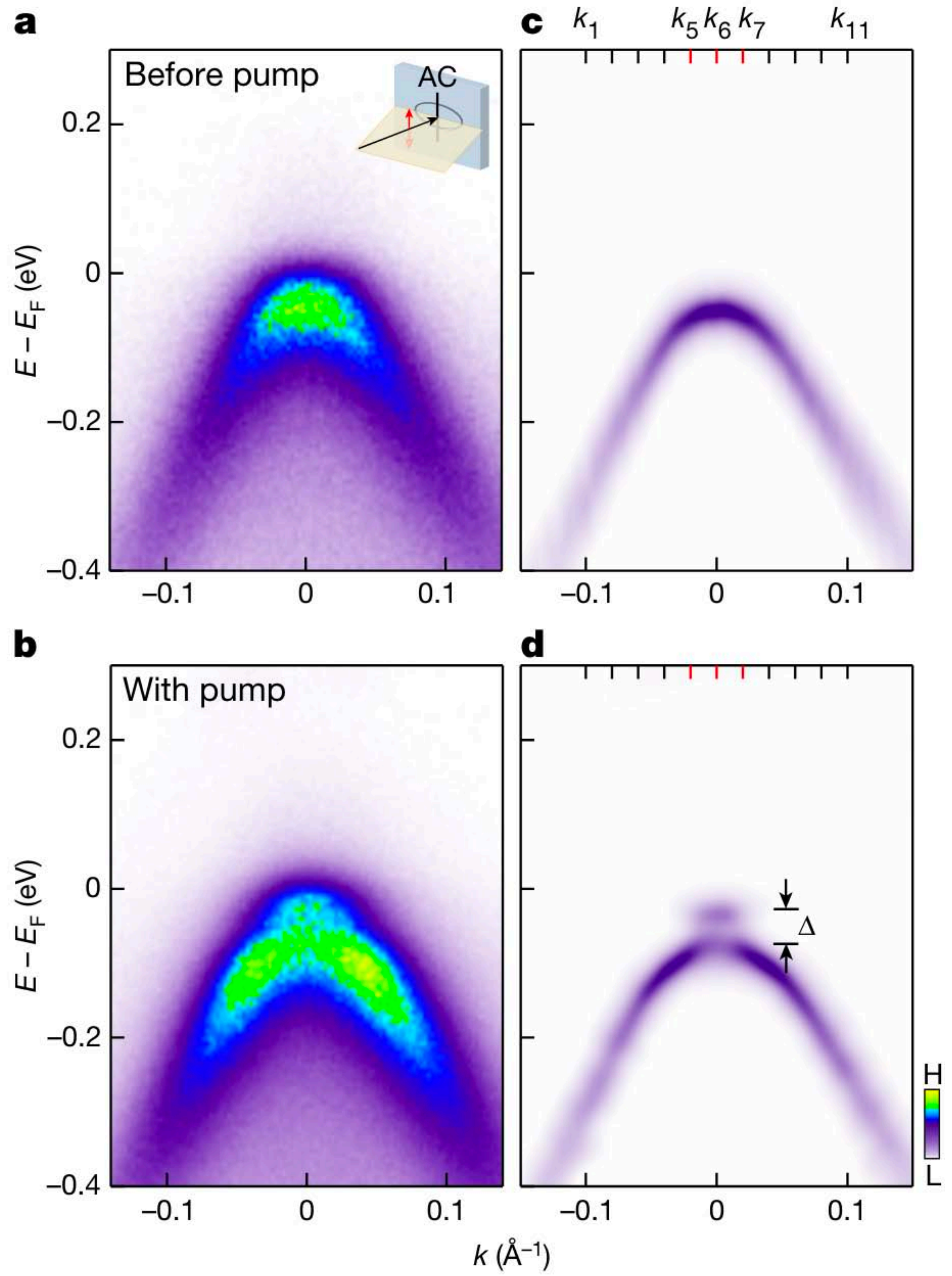
Giant modulation of optical nonlinearity by Floquet engineering



J.-Y. Shan, et int., David Hsieh, Nature 600, 235 (2021)

Black phosphorus

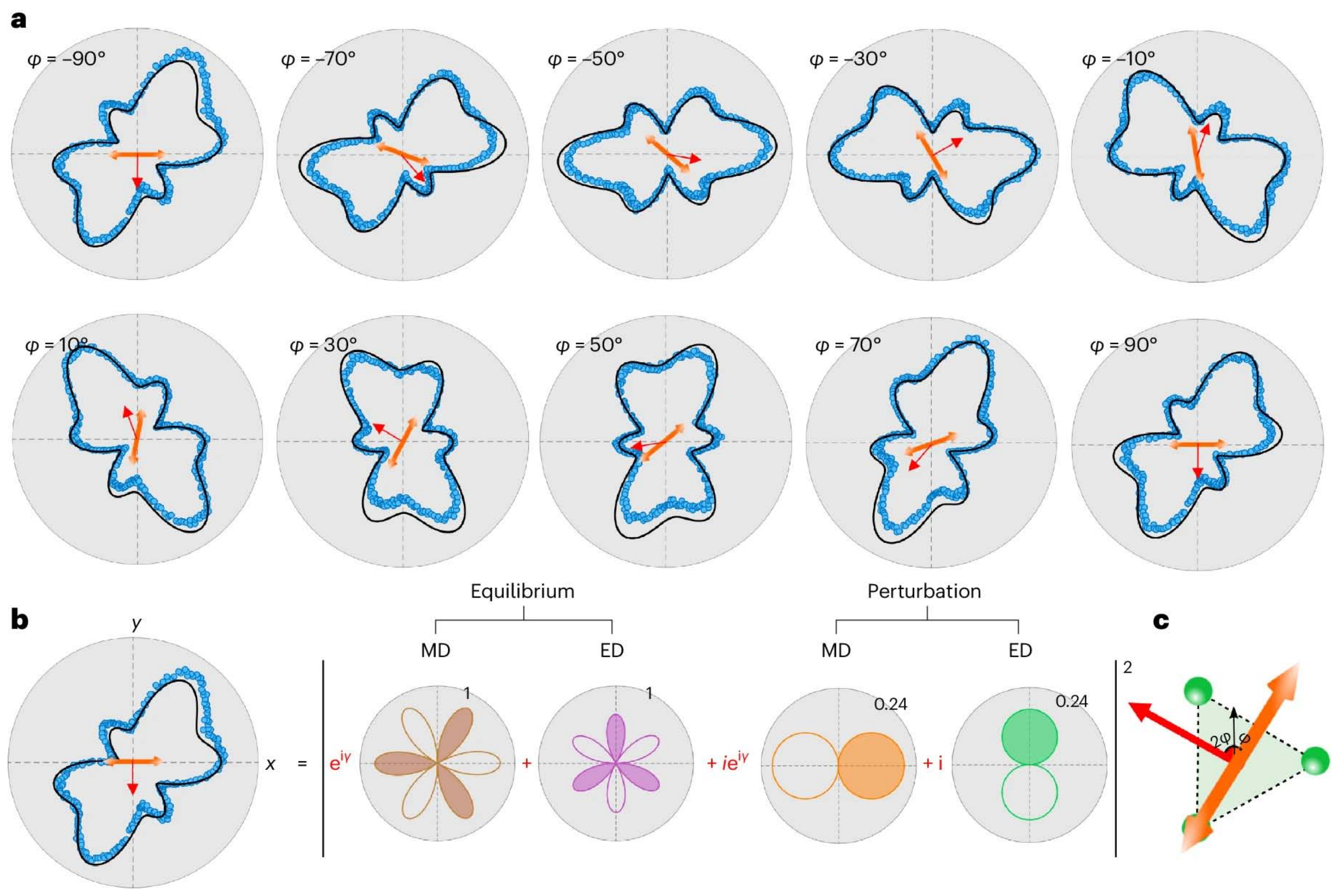
Pseudospin-selective Floquet band engineering in black phosphorus



S. Zhou, et int., Shuyun Zhou, Nature 614, 75 (2023)

Cr2O3

Light-induced electronic polarization in antiferromagnetic Cr_2O_3



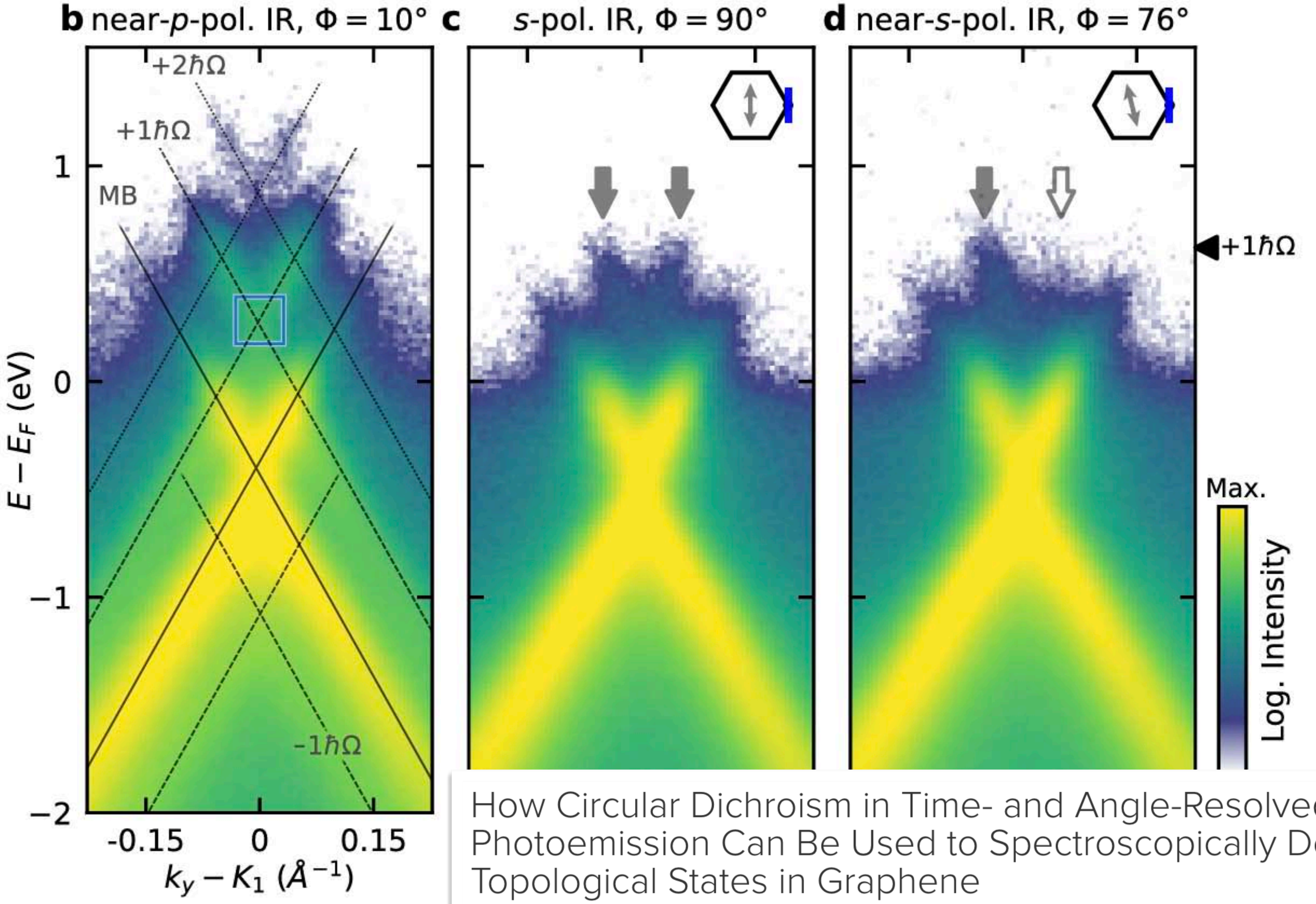
X. Zhang, et int., Anshul Kogar, Nature Materials 2024

… and what about graphene?

Observation of Floquet states in graphene

Marco Merboldt§,[1] Michael Schüler§,[2,3] David Schmitt,[1] Jan Philipp Bange,[1] Wiebke Bennecke,[1] Karun Gadge,[4] Klaus Pierz,[5] Hans Werner Schumacher,[5] Davood Momeni,[5] Daniel Steil,[1] Salvatore R. Manmana,[4] Michael A. Sentef,[6,7,*] Marcel Reutzel,[1,†] and Stefan Mathias[1,8,‡]

Floquet states in graphene interfere with Volkov states

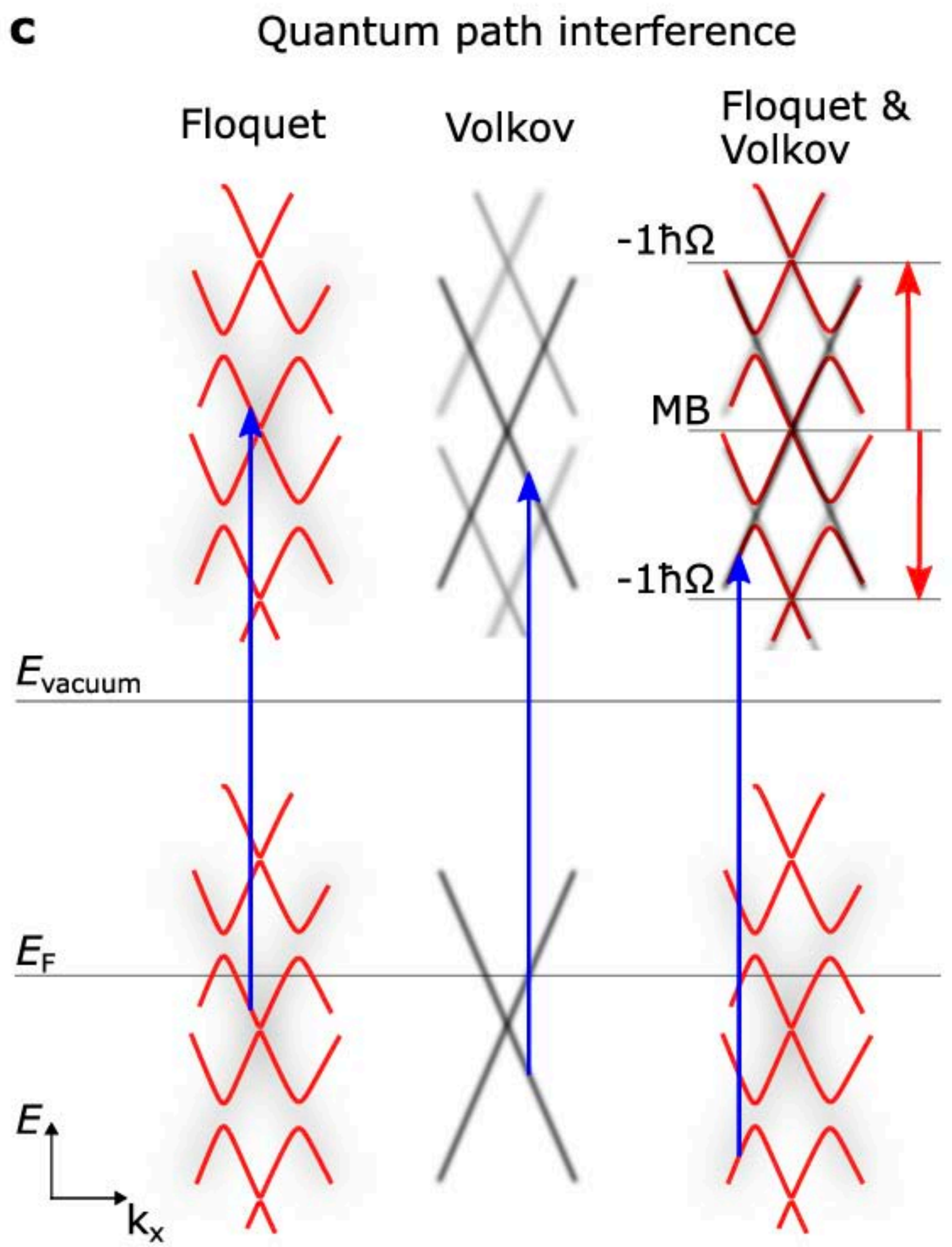


How Circular Dichroism in Time- and Angle-Resolved Photoemission Can Be Used to Spectroscopically Detect Transient Topological States in Graphene

Michael Schüler, Umberto De Giovannini, Hannes Hübener, Angel Rubio, Michael A. Sentef, Thomas P. Devereaux, and Philipp Werner

Phys. Rev. X **10**, 041013 – Published 19 October 2020

outstanding
Floquet topology under circularly polarized light



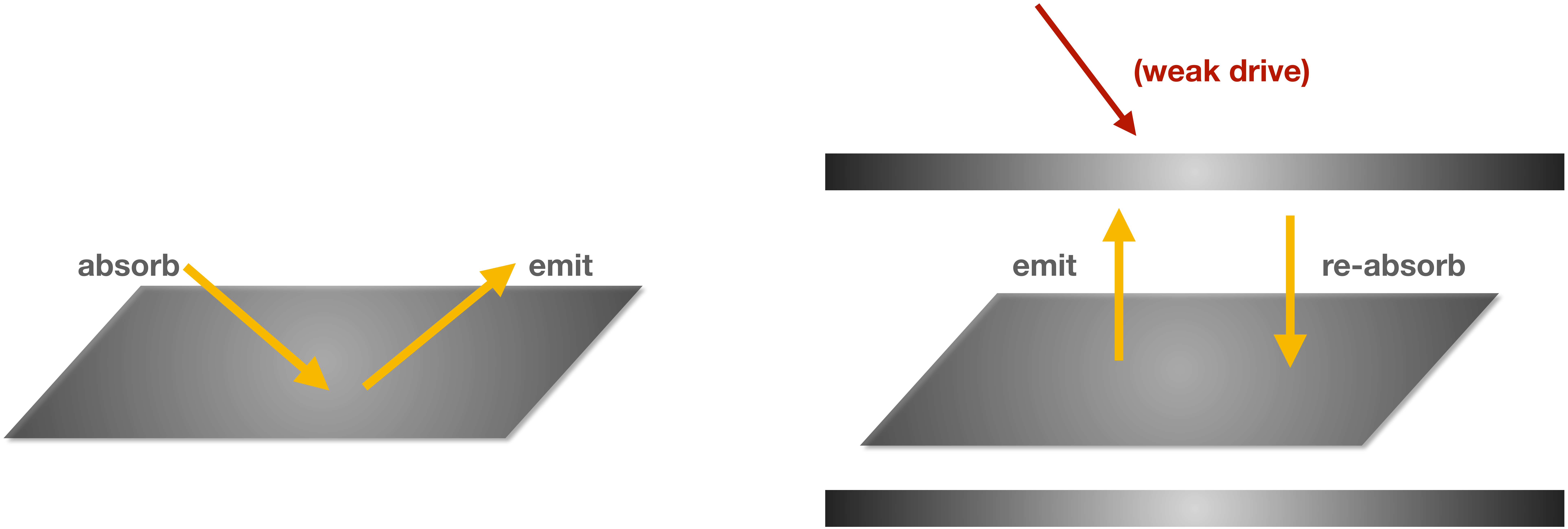
also see Choi, et int., Gedik, arXiv:2404.14392

Outline

Floquet basics
Floquet in solids
→ Floquet to cavity

Going beyond classical drives — cavity quantum materials

Replacing strong fields by strong coupling



best of both worlds: flexibility of driven systems + efficiency of cavities

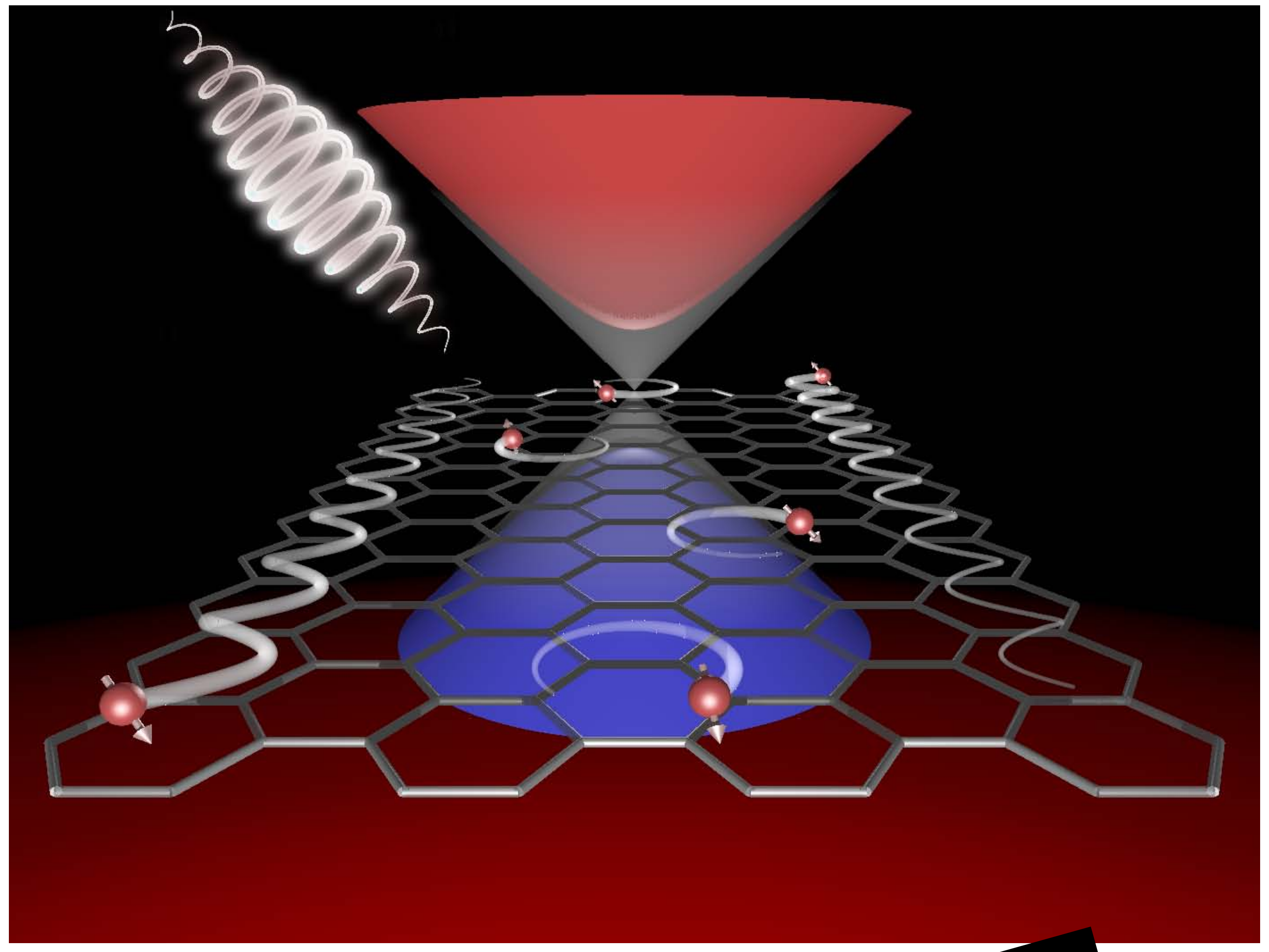
Schlawin, Kennes, Sentef
Applied Physics Reviews 9, 011312 (2022)

also recommended:
Bloch, Cavalleri, Galitski, Hafezi, Rubio, Nature 606, 41 (2022)

Back to Haldane: Floquet engineering without (real) photons

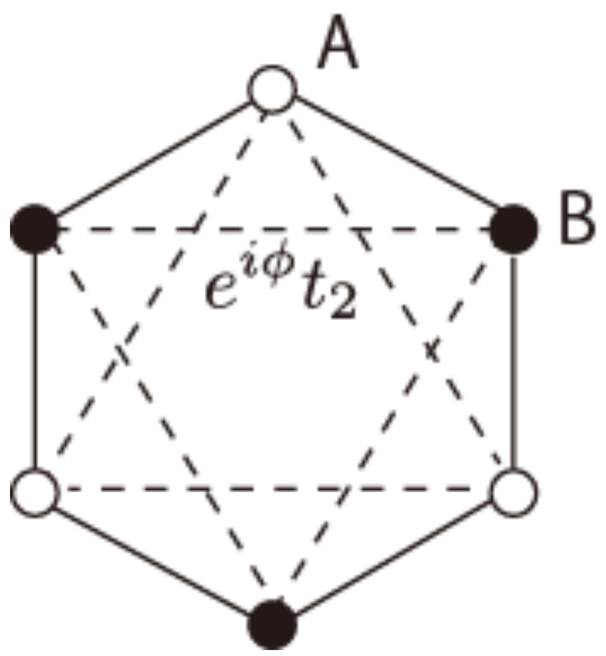
Quantum Hall effect without a magnetic field

Graphene + circularly polarized light
(breaks time reversal)



?

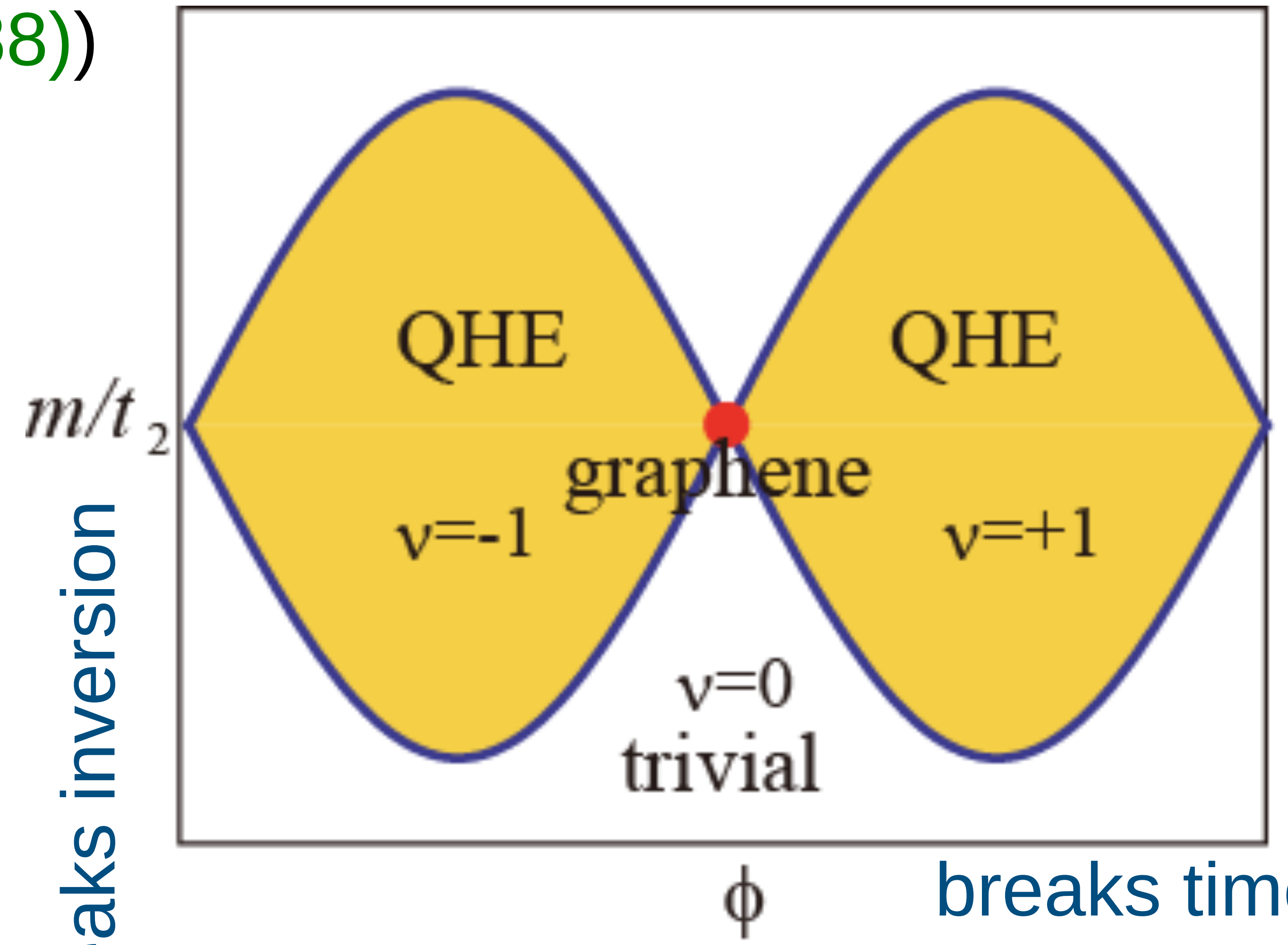
Haldane model (PRL 61, 2015 (1988))



Local flux ϕ

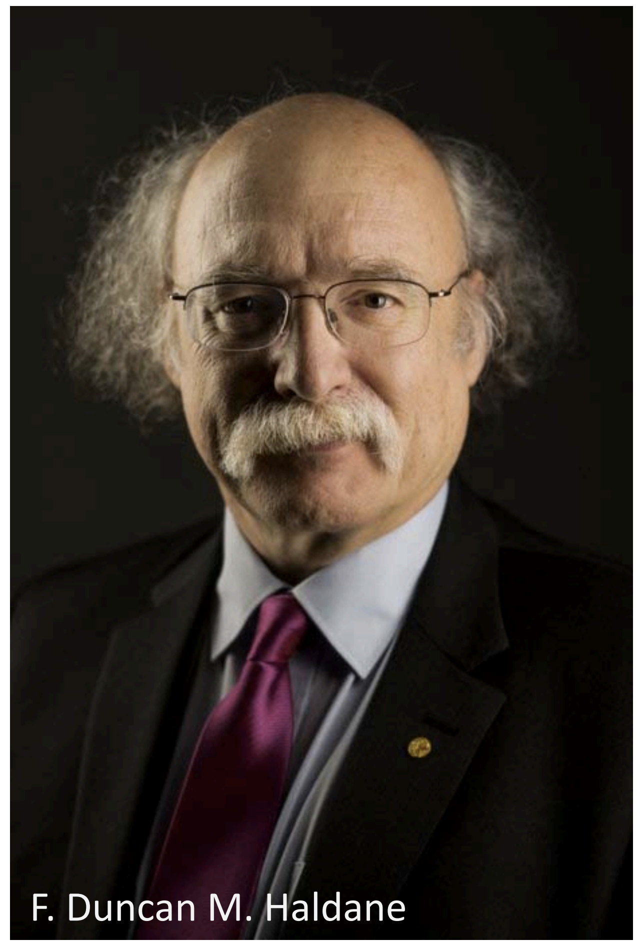
Staggered field $m = E_A - E_B$

Fictitious fields!



breaks inversion

breaks time reversal



(c) Nobel Media AB, Photo: A. Mahmoud

~~Need concept of Floquet engineering~~

Need concept of cavity engineering

Cavity Chern insulator

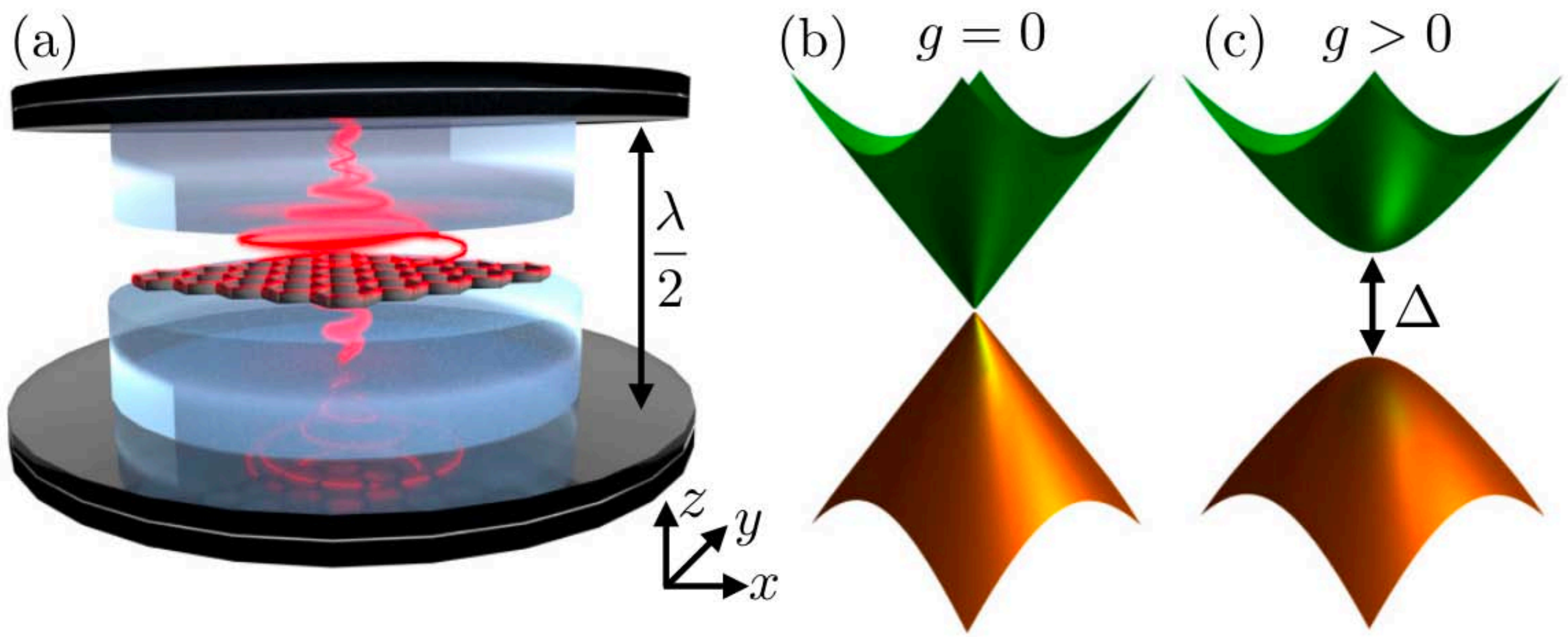
Goal: induce Haldane mass term by coupling to cavity with circularly polarized photon mode

PHYSICAL REVIEW B **99**, 235156 (2019)

Cavity quantum electrodynamical Chern insulator: Towards light-induced quantized anomalous Hall effect in graphene

Xiao Wang,[1] Enrico Ronca,[2] and Michael A. Sentef[2,*]

also cf. Kibis, Kyriienko and Shelykh, PRB 84, 195413 (2011)



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

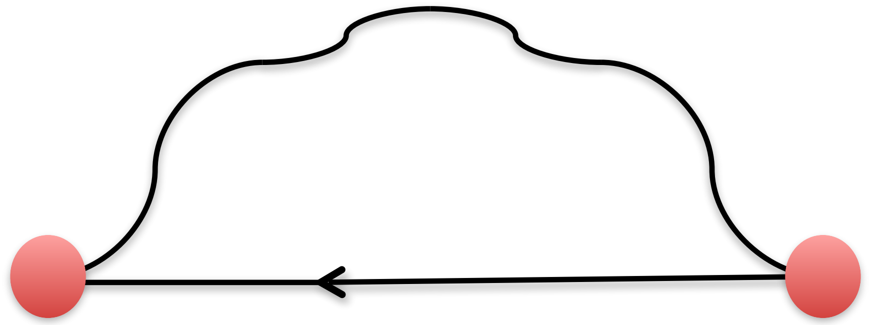
$$\hat{\vec{A}} = A_0 \sum_\lambda (\vec{e}_\lambda a_\lambda + \vec{e}^*_\lambda a^\dagger_\lambda)$$

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

$$\gamma(\vec{k}) = \hbar v_F(k_x + ik_y)$$

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

effective electron-photon coupling strength $g \equiv v_F A_0 \sqrt{2}$



compute electronic self-energy (2x2) due to el-phot coupling (lowest order in g):

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

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

Matsubara summation + analytical continuation:

$$\Sigma^R_{0,aa}(\vec{k}, \epsilon) = -g^2 \frac{(\epsilon + i0^+ - \omega)n_B(-\omega) + \frac{1}{2}[(\epsilon + i0^+ - \omega + v_F k)n_F(v_F k) + (\epsilon + i0^+ - \omega - v_F k)(1 - n_F(v_F k))]}{(\epsilon + i0^+ - \omega)^2 - v_F^2 k^2}$$

$$\Sigma^R_{0,bb}(\vec{k}, \epsilon) = g^2 \frac{(\epsilon + i0^+ + \omega)n_B(\omega) + \frac{1}{2}[(\epsilon + i0^+ + \omega + v_F k)n_F(v_F k) + (\epsilon + i0^+ + \omega - v_F k)(1 - n_F(v_F k))]}{(\epsilon + i0^+ + \omega)^2 - v_F^2 k^2}.$$

Cavity Chern insulator

At Dirac point k=0:

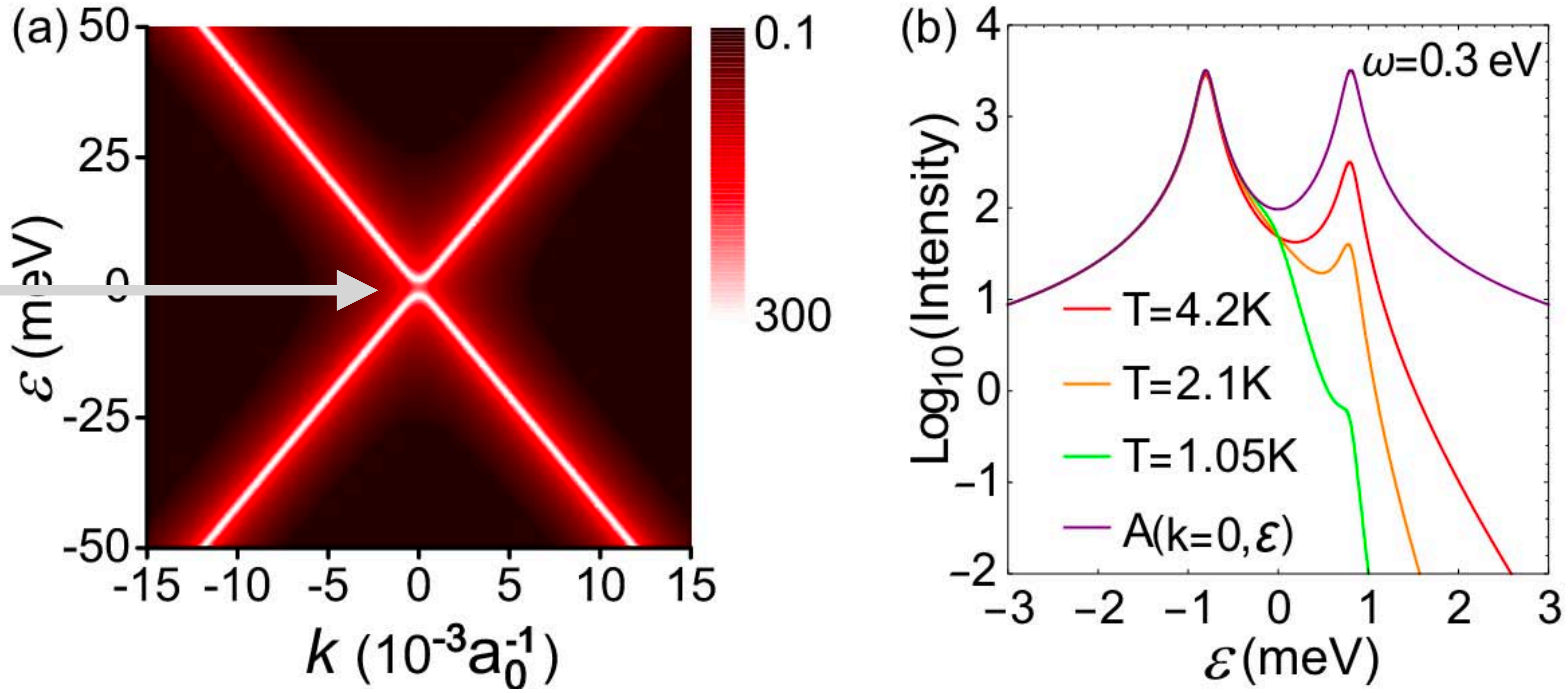
$$\Sigma^R_{0,aa}(0,\epsilon) = -g^2 \frac{(\epsilon + i0^+ - \omega)(-1) + \frac{1}{2}(\epsilon + i0^+ - \omega)}{(\epsilon + i0^+ - \omega)^2}$$

$$= \frac{g^2/2}{\epsilon + i0^+ - \omega}, \quad \text{(A26)}$$

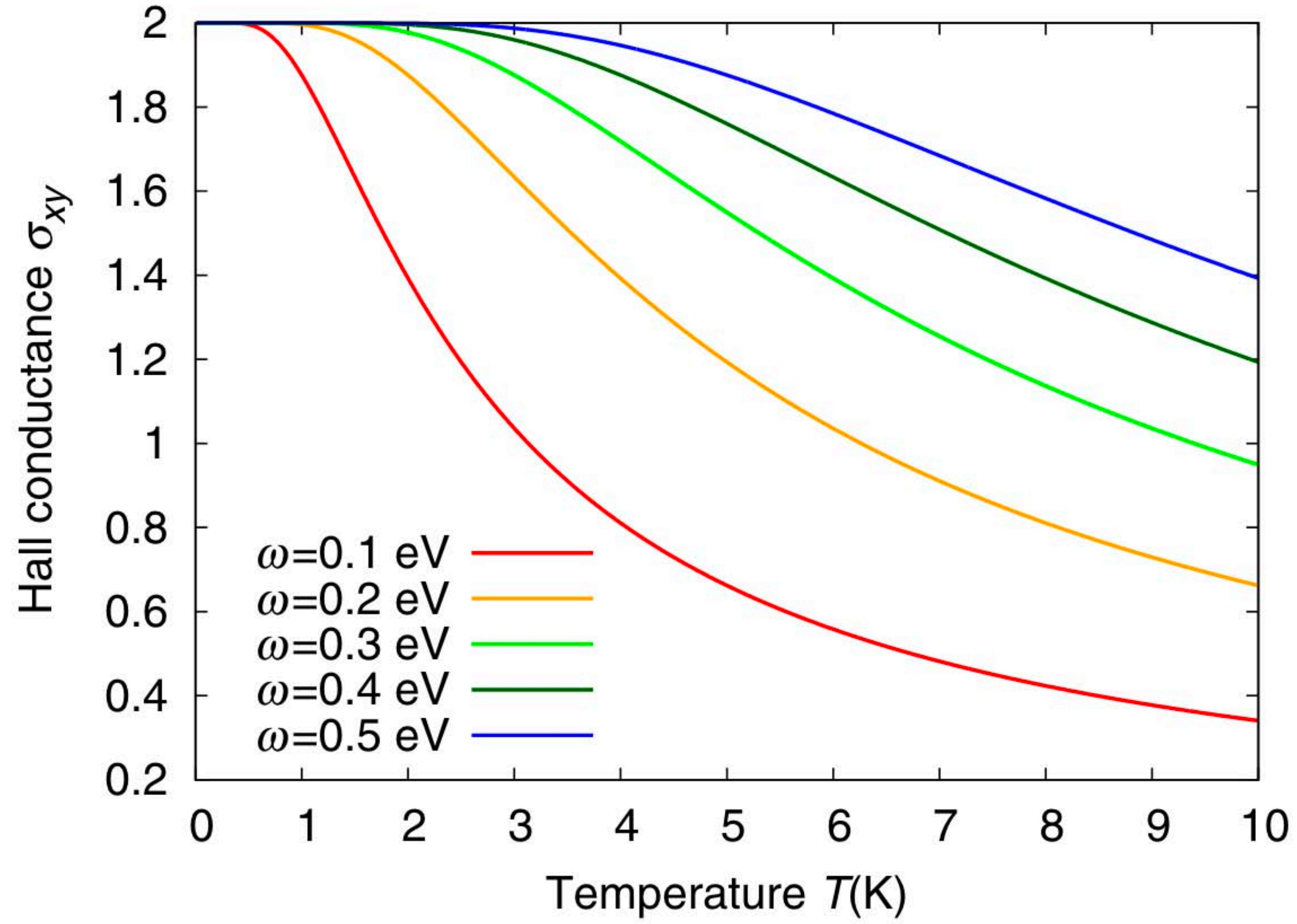
$$\Sigma^R_{0,bb}(0,\epsilon) = g^2 \frac{\frac{1}{2}(\epsilon + i0^+ + \omega)}{(\epsilon + i0^+ + \omega)^2} = \frac{g^2/2}{\epsilon + i0^+ + \omega}. \quad \text{(A27)}$$

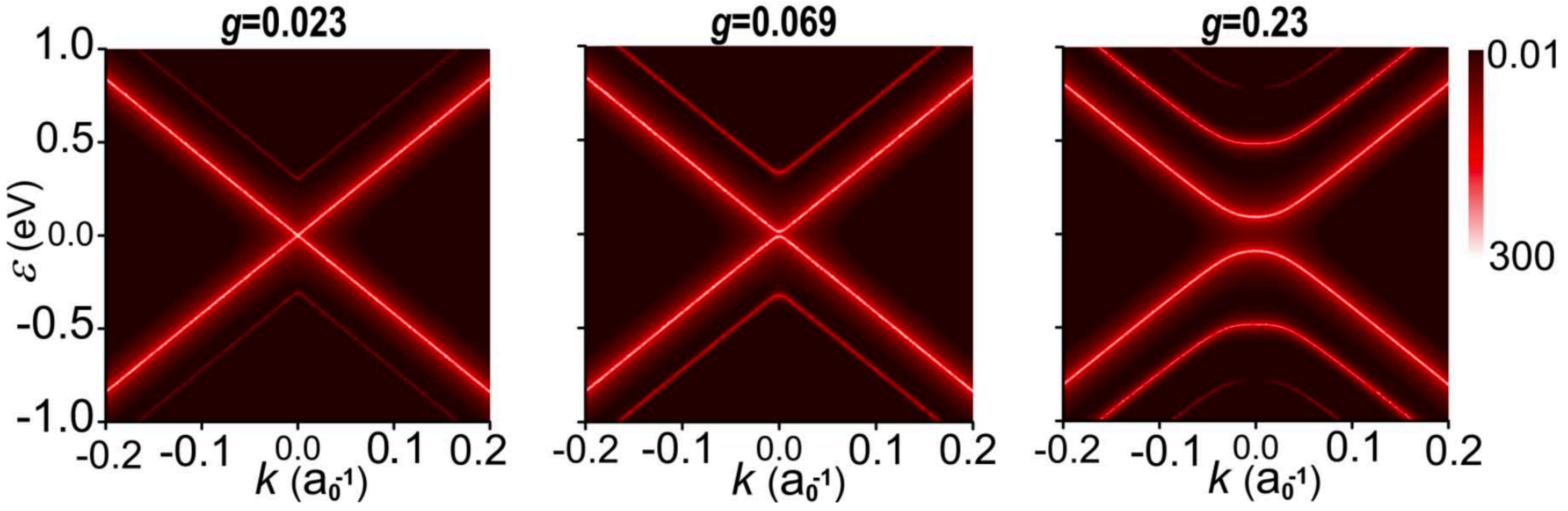
Considering the lowest-order self-energy at the Dirac point $\hat{\Sigma}^R(k=0,\epsilon) \to \hat{\Sigma}^R_0(k=0,\epsilon)$ from Eqs. (3) and (4), we find that $A(k=0,\epsilon)$ acquires an energy gap $\Delta = \sqrt{2g^2 + \omega^2} - \omega$. In the limit $2g^2/\omega^2 \ll 1$, we obtain $\Delta \approx \frac{g^2}{\omega} = \frac{2\hbar^2 v_F^2 A_0^2}{\omega}$. This result is in remarkably close formal analogy with the Floquet high-frequency expansion [14] when the quantum photon amplitude A_0 is replaced by the field strength A_0 of the classical vector potential.

Haldane gap



Quantized cavity Hall effect at suffiently low T



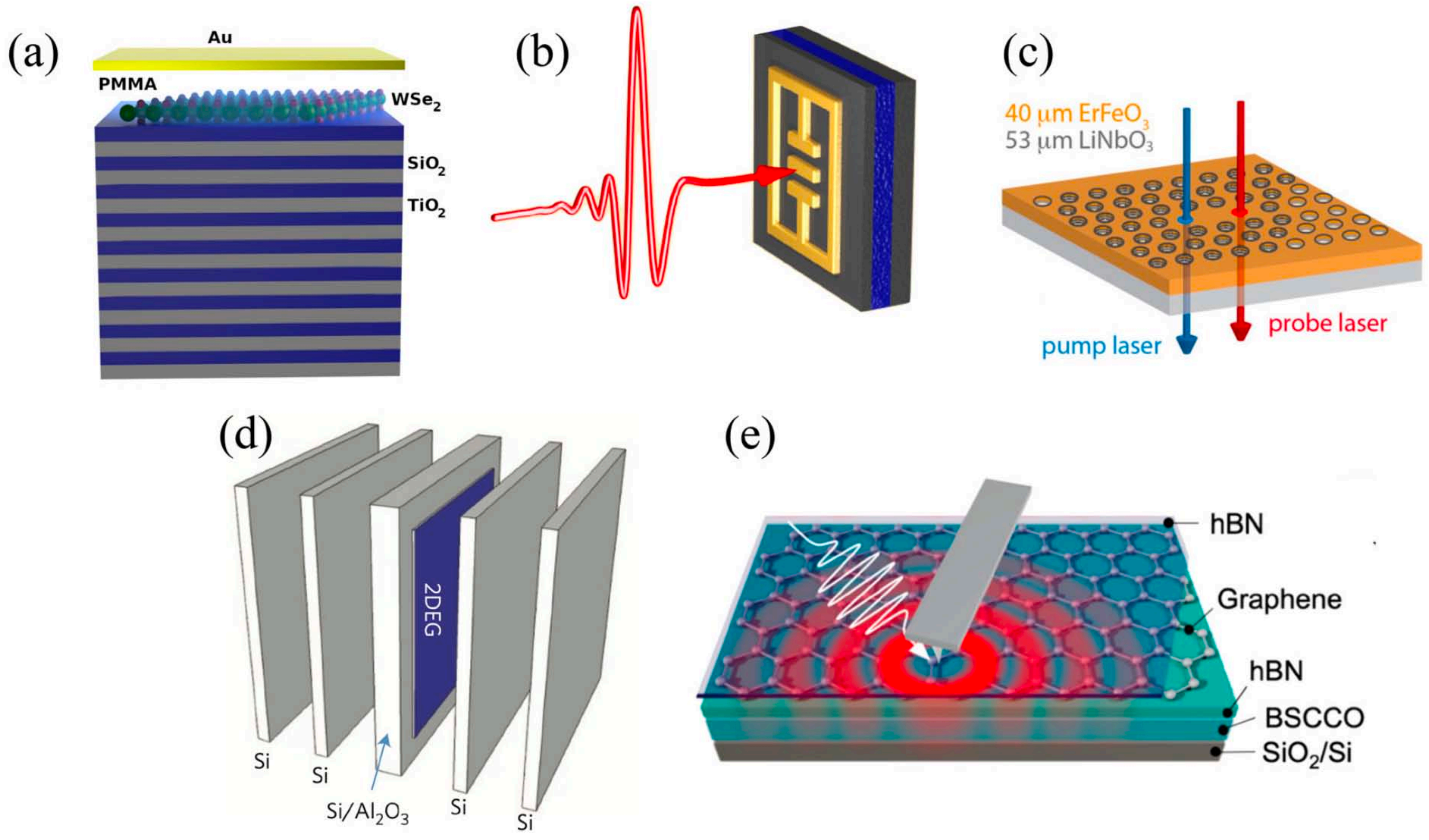


(self-consistent results with analytical continuation via Padé)

Going beyond classical drives — cavity quantum materials

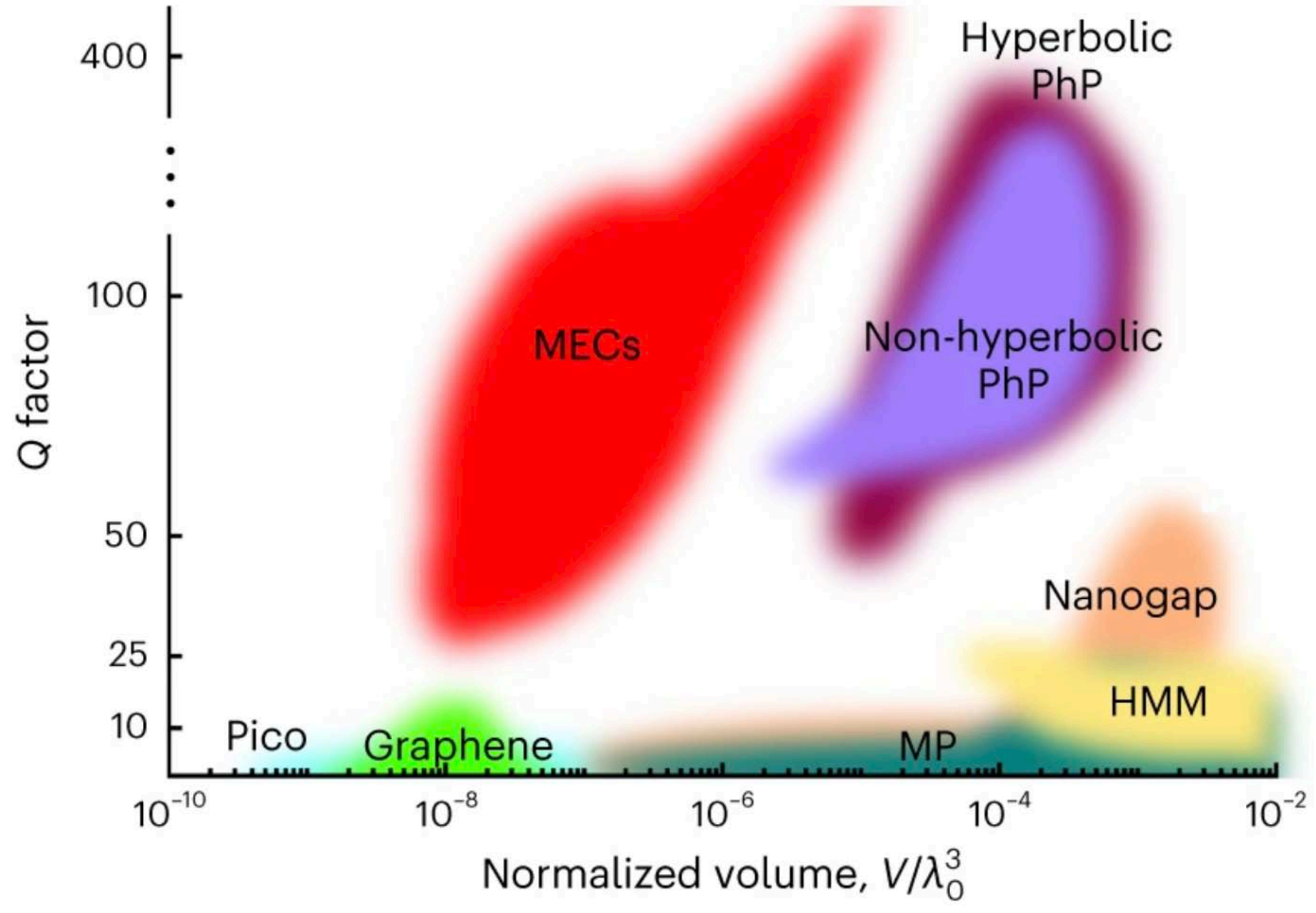
Replacing strong fields by strong coupling

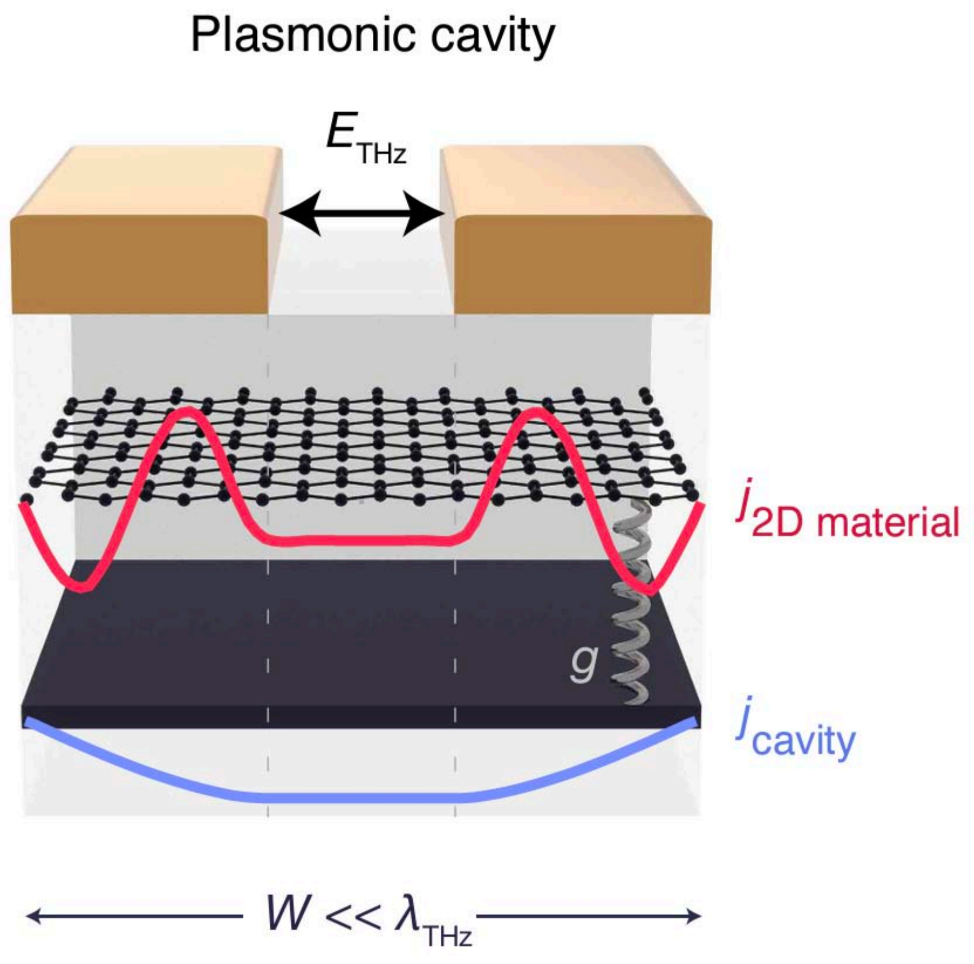
Many possible platforms (not just Fabry-Perot)



e.g., near-field effects on surfaces

Herzig Sheinfux et al., Nat. Mater. 23, 499 (2024)



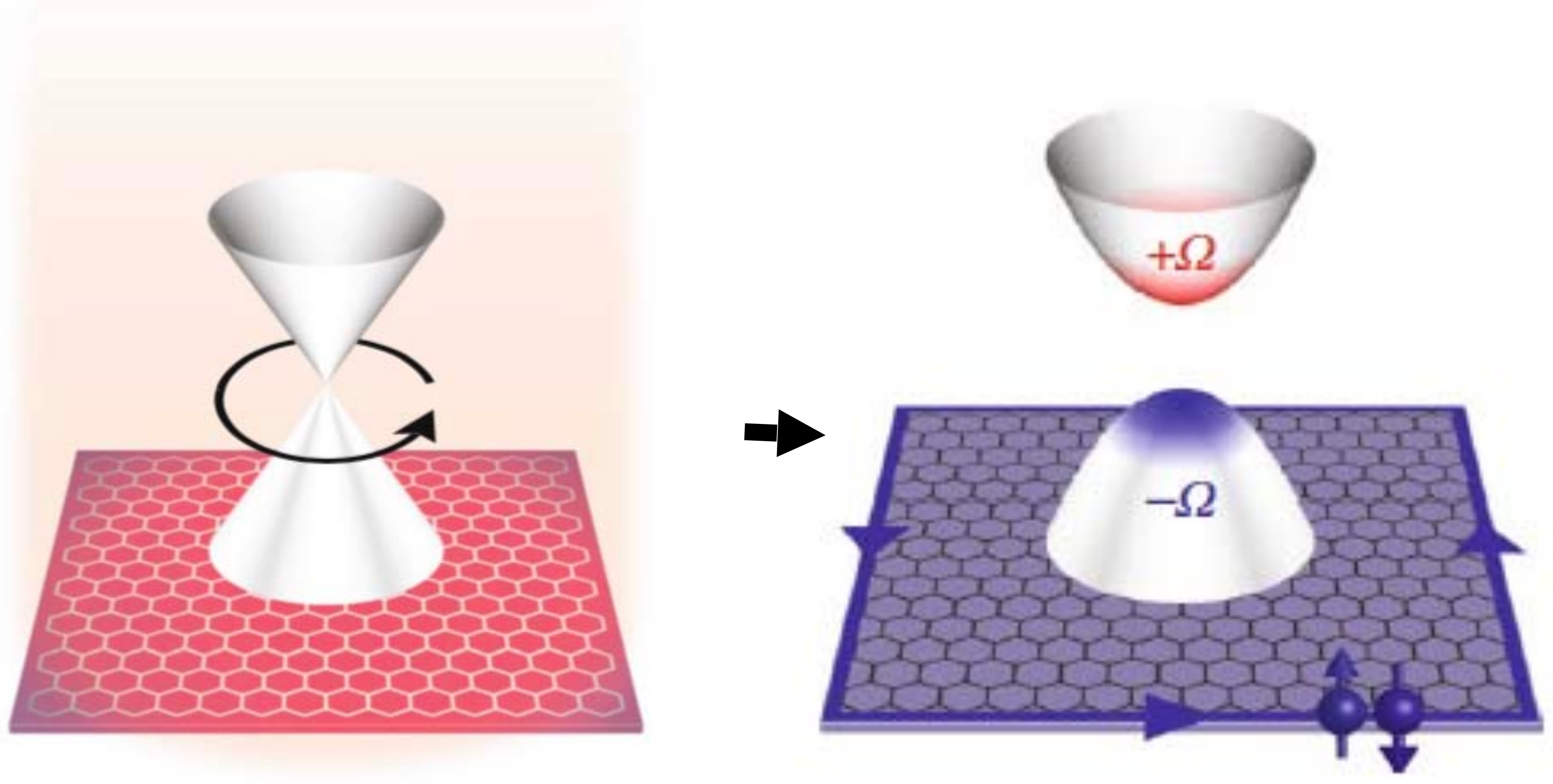


Schlawin, Kennes, Sentef
[Applied Physics Reviews 9, 011312 (2022)]

also recommended:
Bloch, Cavalleri, Galitski, Hafezi, Rubio, Nature 606, 41 (2022)

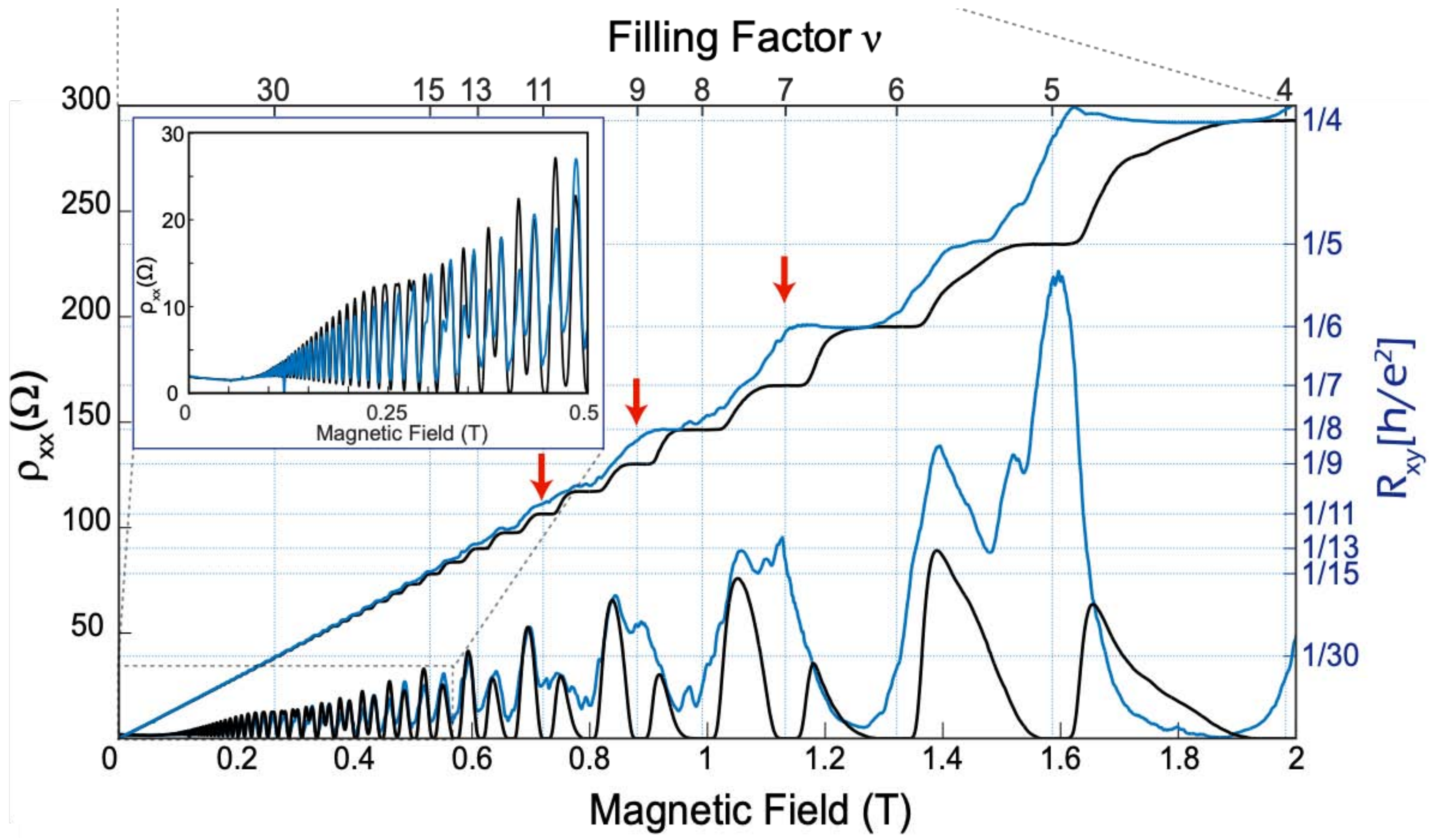
Light-matter control of quantum materials: classical to quantum light

light-induced topological matter



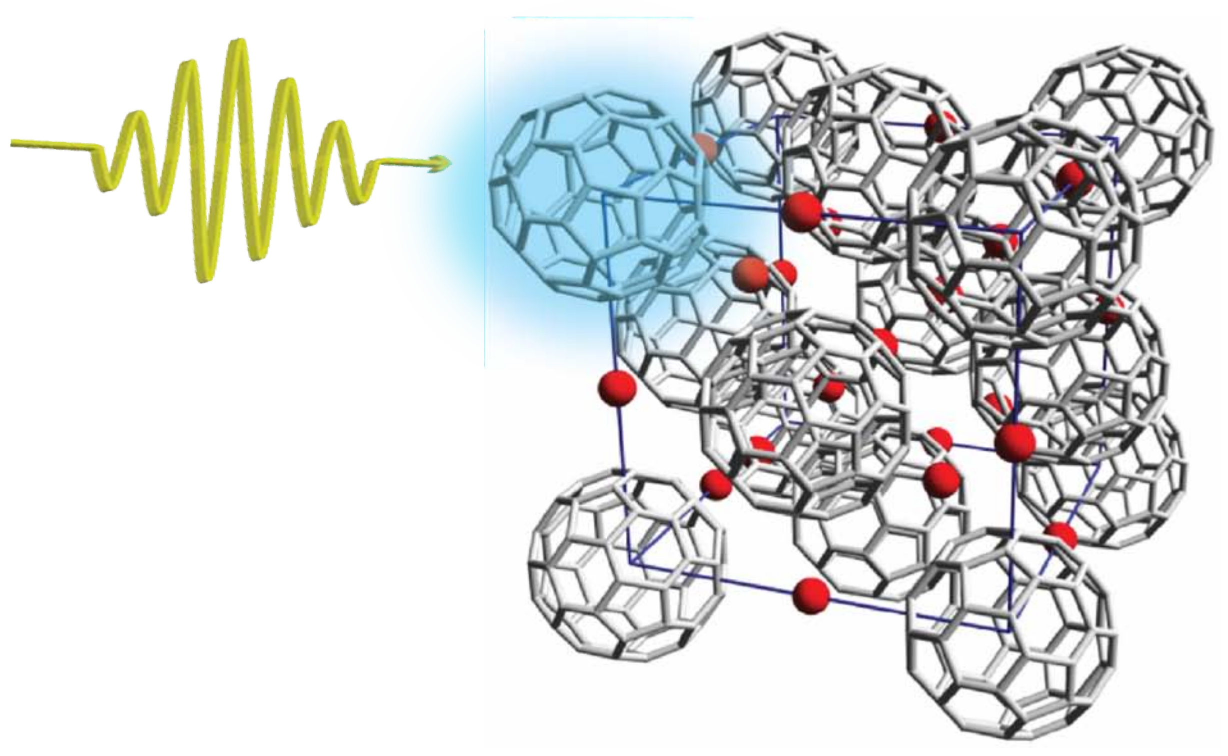
Oka and Aoki, PRB 79, 081406 (2009)
McIver et al., Nat. Phys. 16, 38 (2020)

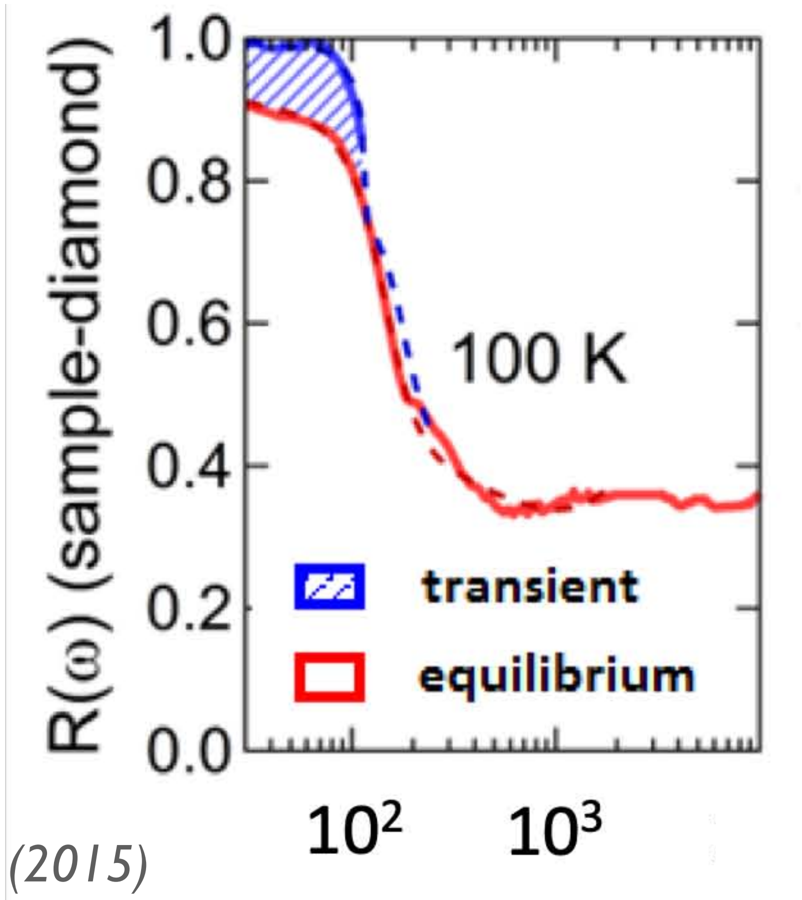
cavity versus topology



Appugliese et al., Science 375, 1030 (2022)

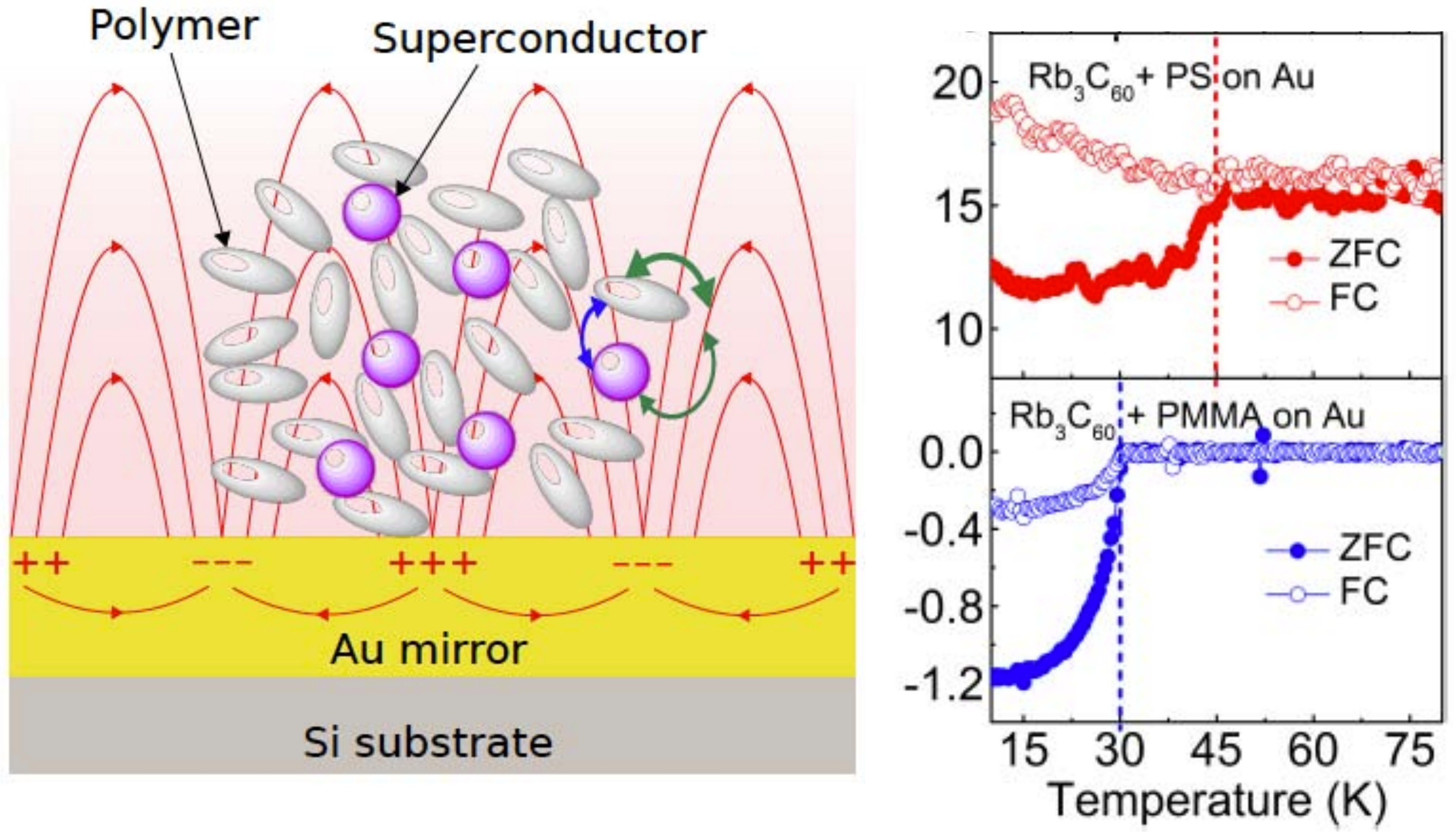
light-induced superconductivity





Mitrano et al., Nature 530, 461 (2015)
Budden et al., Nat. Phys. 17, 611 (2021)

cavity-induced superconductivity?



Thomas et al., arXiv:1911.01459; Nano Lett. 21, 4365 (2021)

Basov, Averitt, Hsieh, Nat. Mater. 16, 1077 (2017)
de la Torre et al., Rev. Mod. Phys. 93, 041002 (2021)

Schlawin, Kennes, Sentef, Applied Physics Reviews 9, 011312 (2022)
Bloch et al., Nature 606, 41 (2022)

Light-matter control of quantum materials: classical to quantum light

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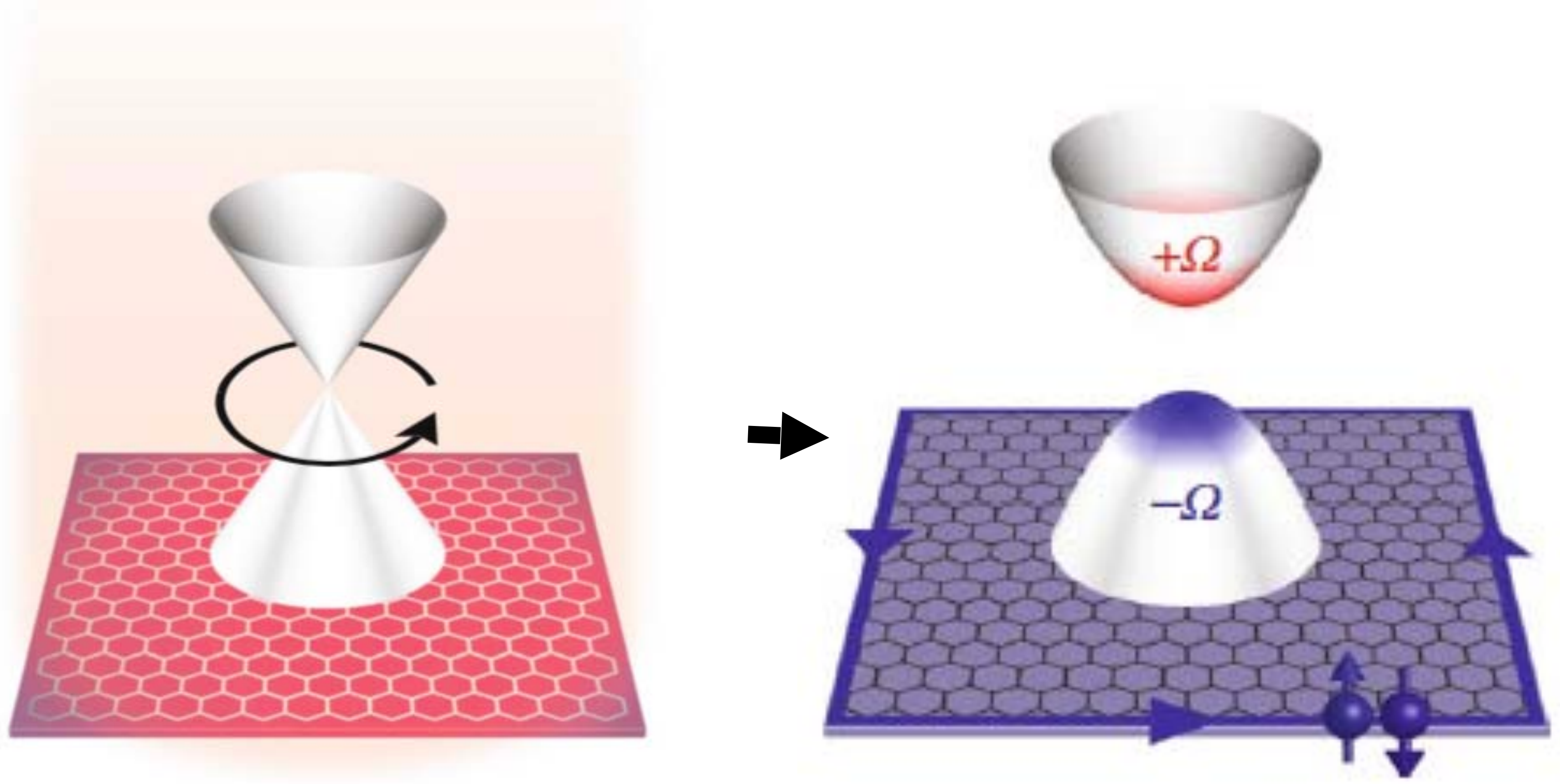
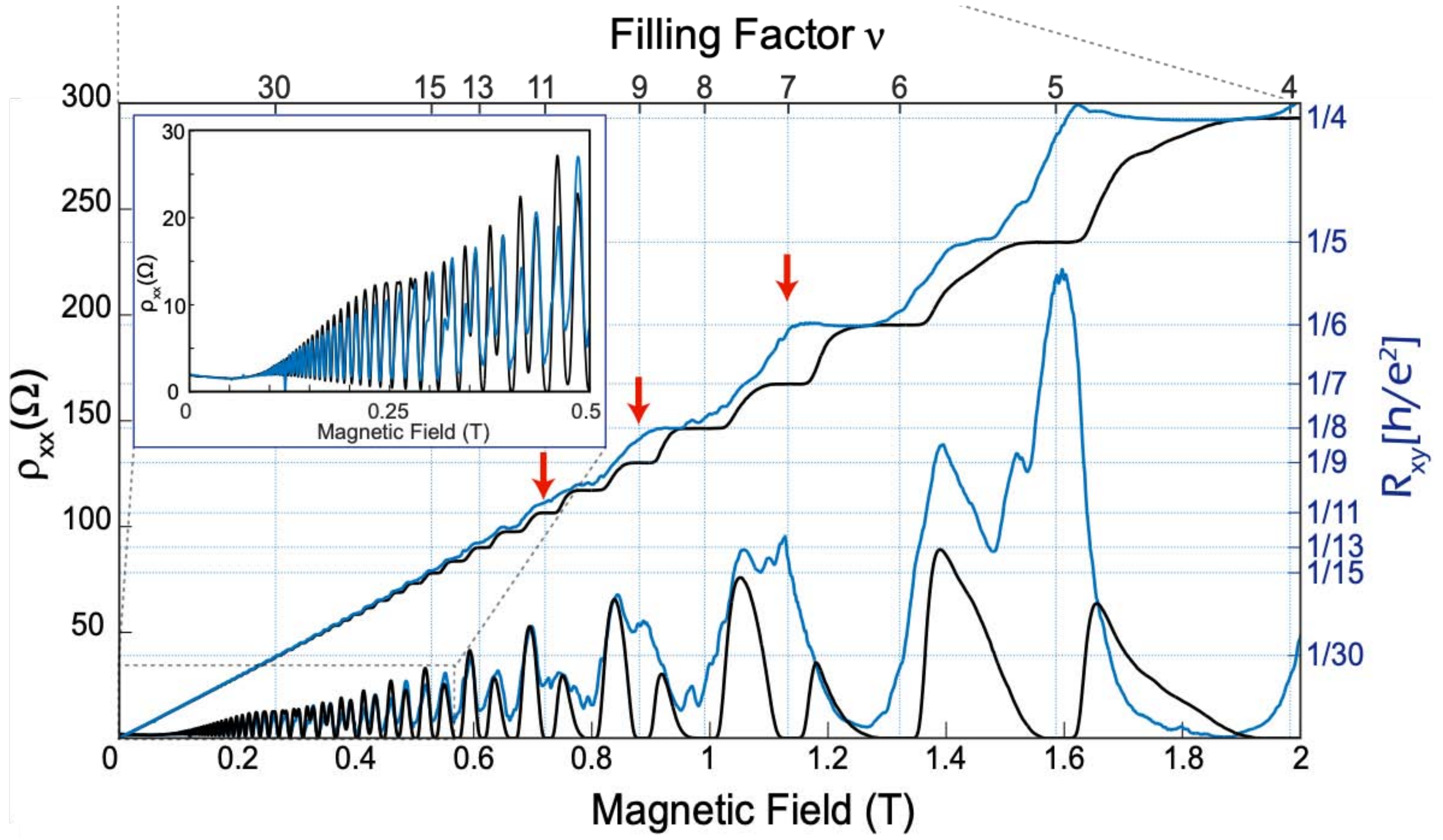


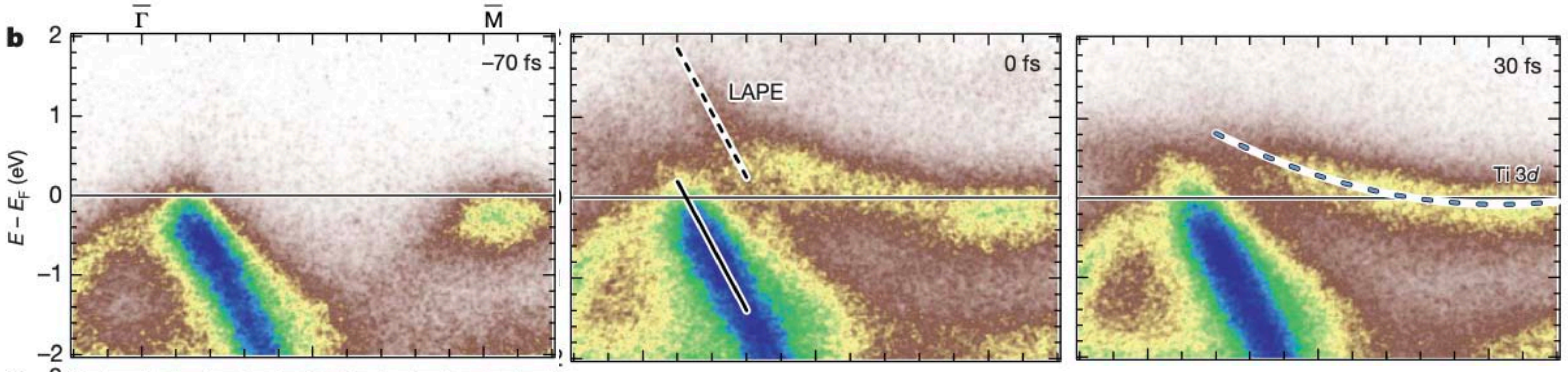
Oka and Aoki, PRB 79, 081406 (2009)
McIver et al., Nat. Phys. 16, 38 (2020)

cavity versus topology



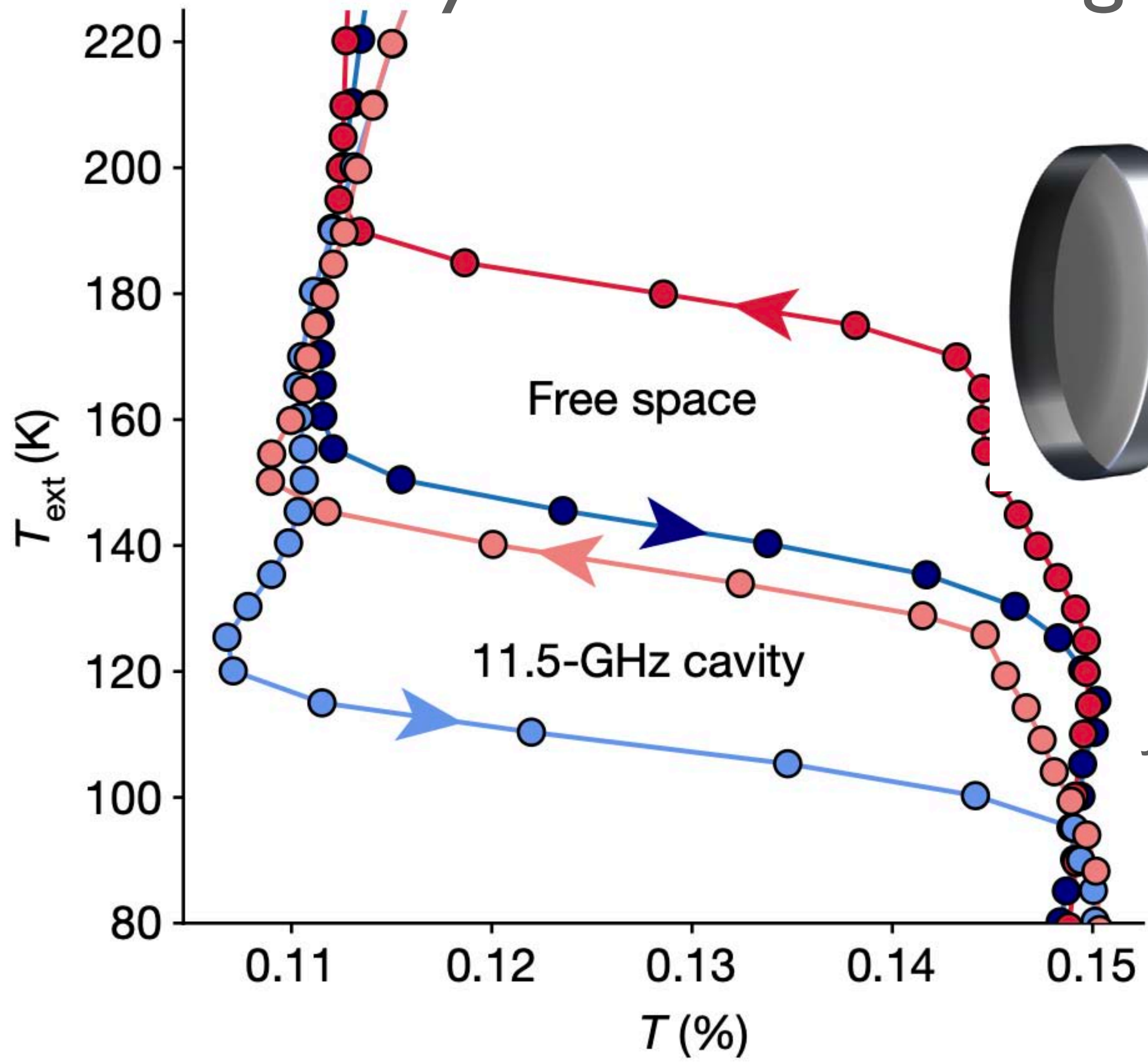
Appugliese et al., Science 375, 1030 (2022)

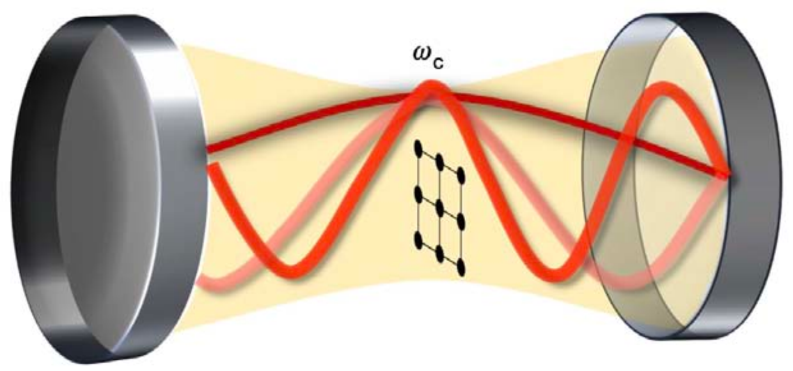
light-driven charge density waves



e.g., Rohwer et al., Nature 471, 490 (2011)

cavity-controlled charge density waves



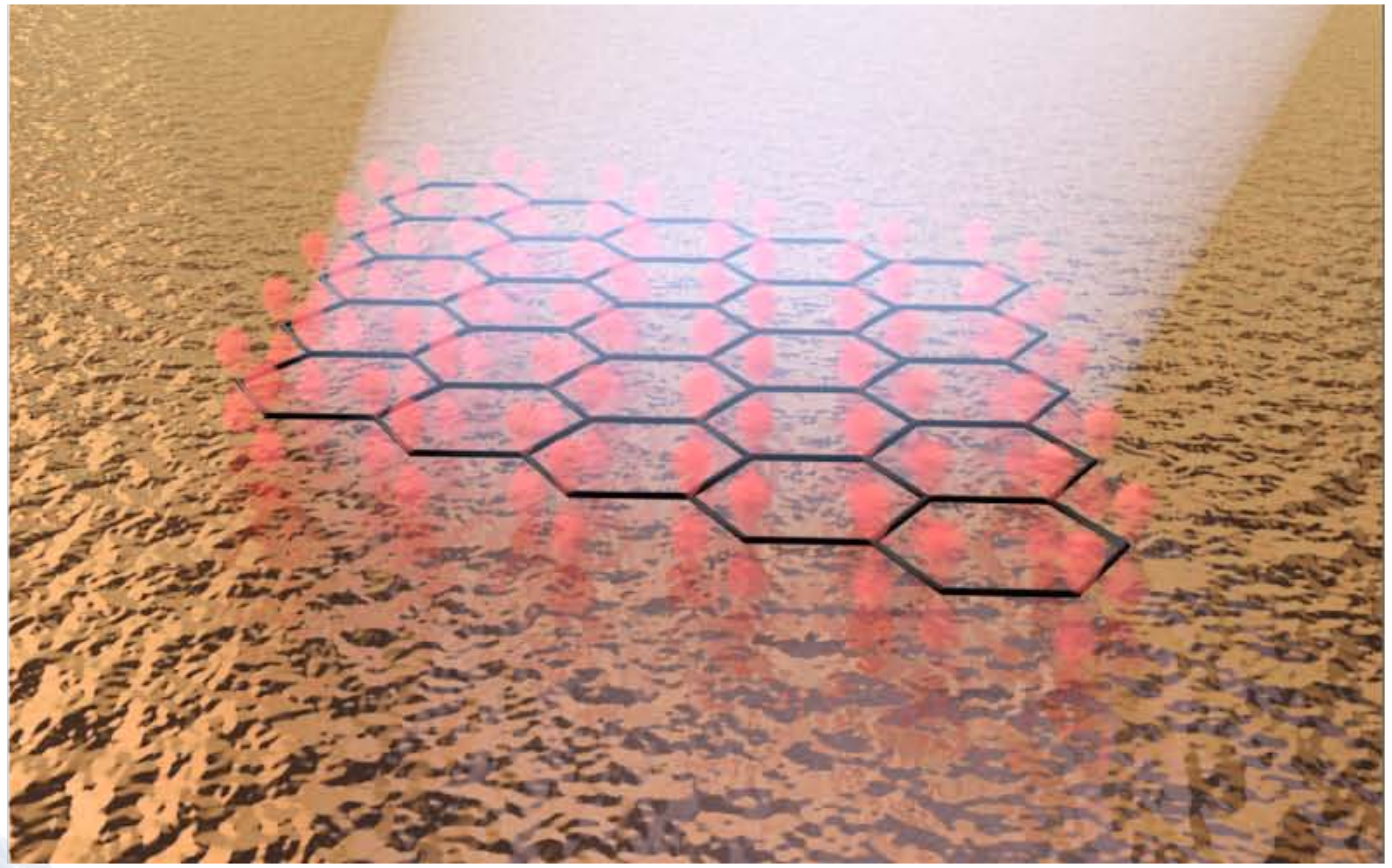


Jarc et al., Nature 622, 487 (2023)

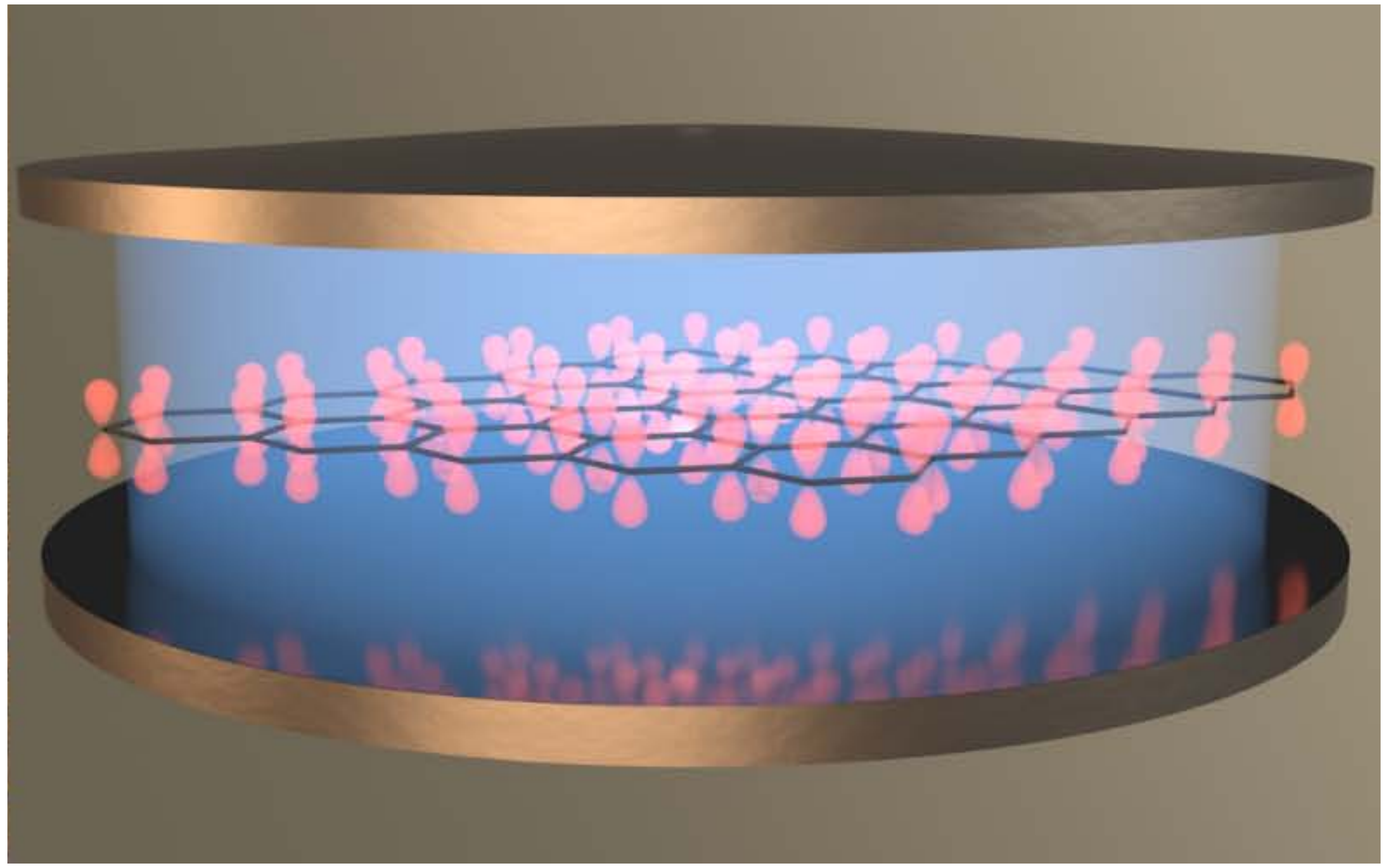
Basov, Averitt, Hsieh, Nat. Mater. 16, 1077 (2017)
de la Torre et al., Rev. Mod. Phys. 93, 041002 (2021)

Schlawin, Kennes, Sentef, Applied Physics Reviews 9, 011312 (2022)
Bloch et al., Nature 606, 41 (2022)

Cavity quantum materials



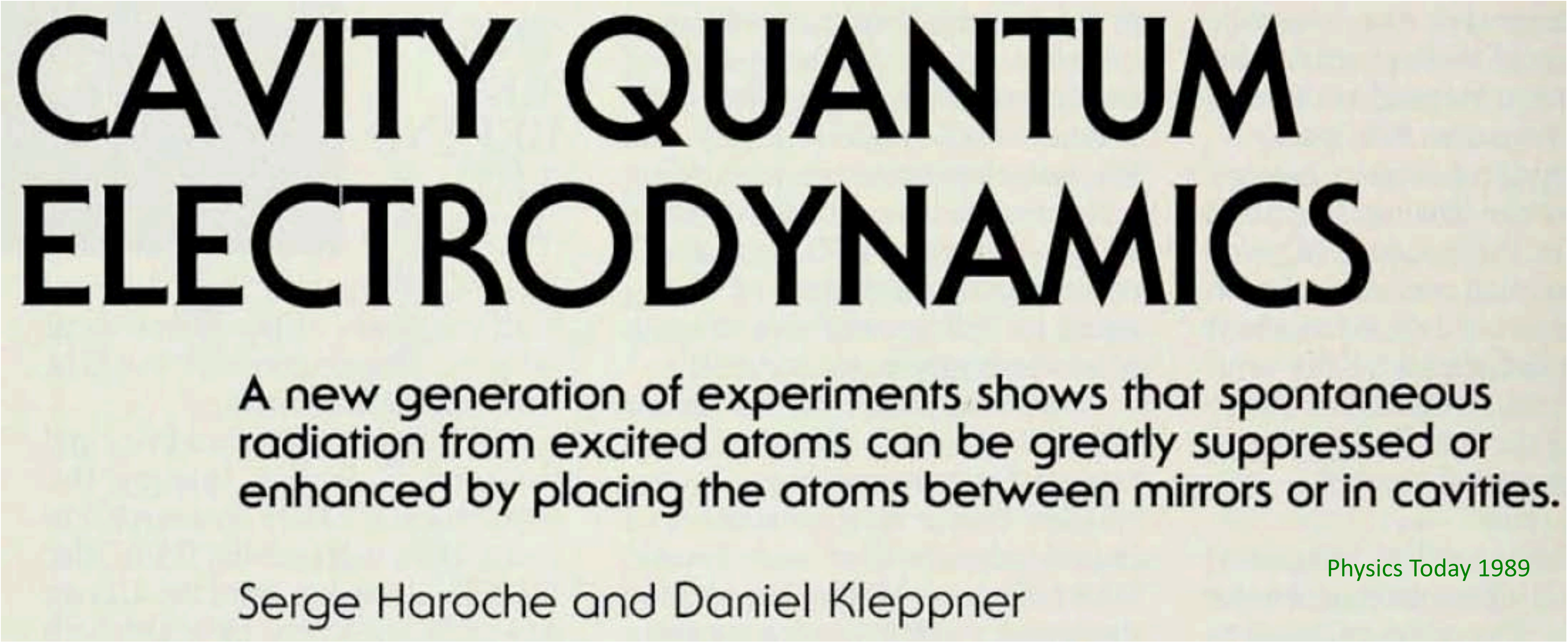
strong laser



strong light-matter coupling

Applied Physics Reviews 9, 011312 (2022)

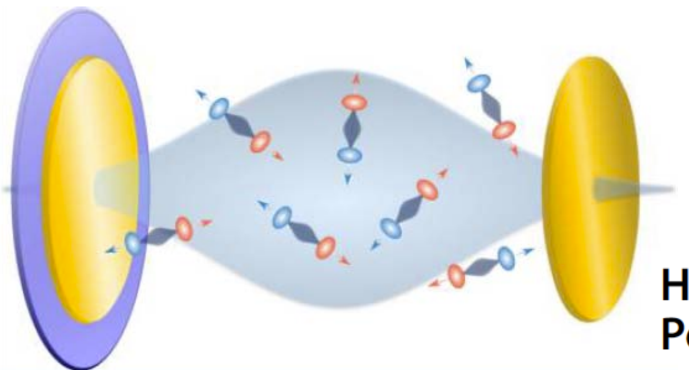
Cavity QED with matter


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

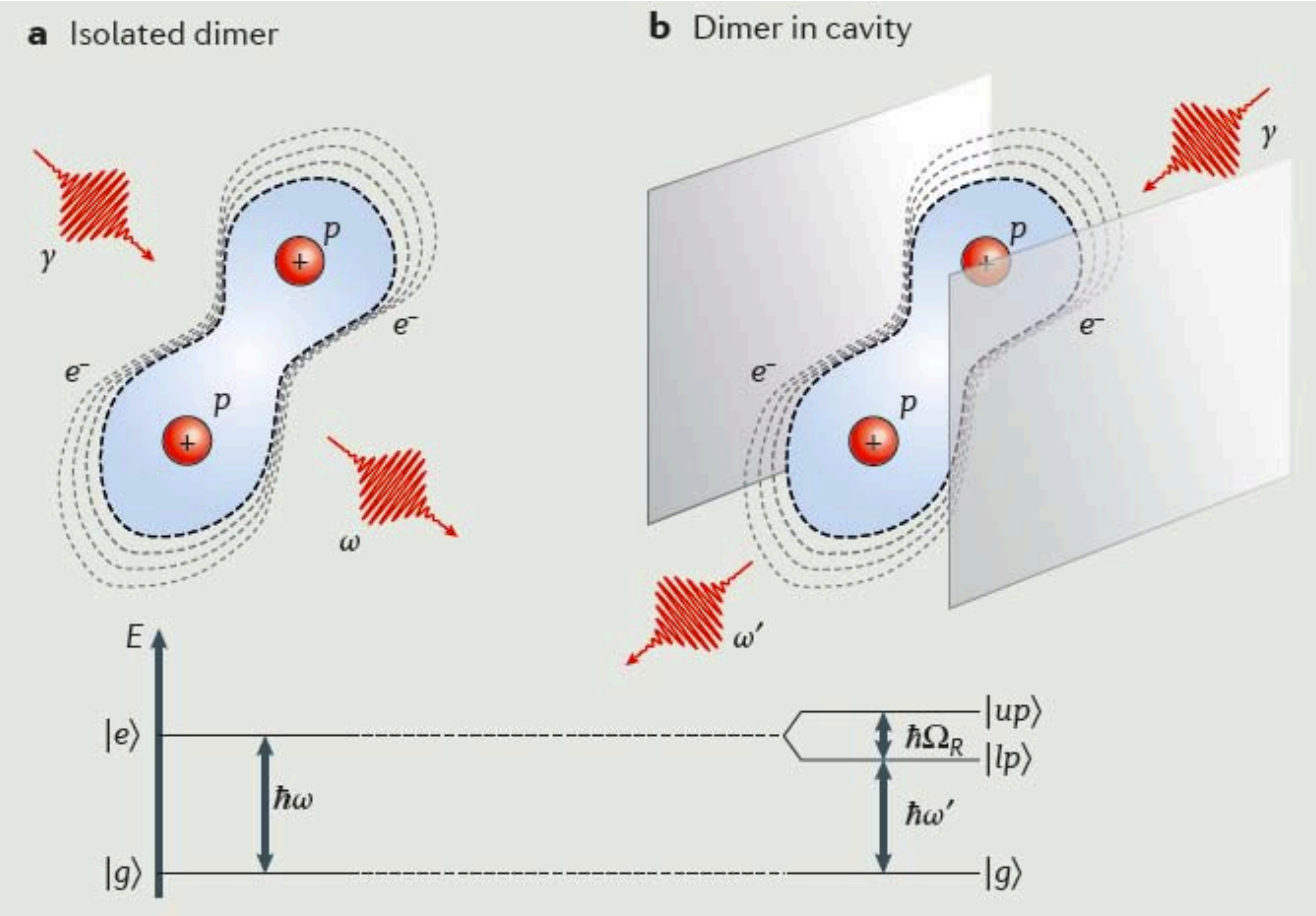


Hybrid Light–Matter States in a Molecular and Material Science Perspective

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

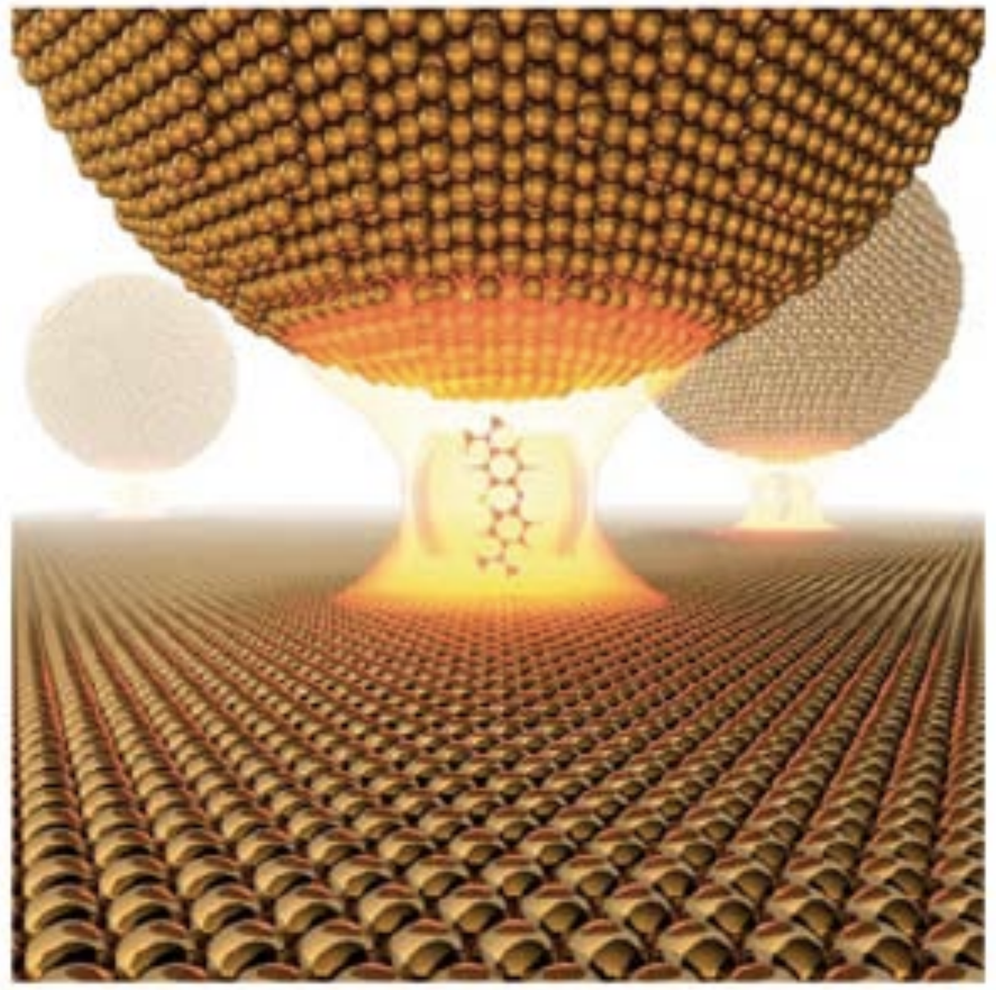
changing the vacuum changes the matter!

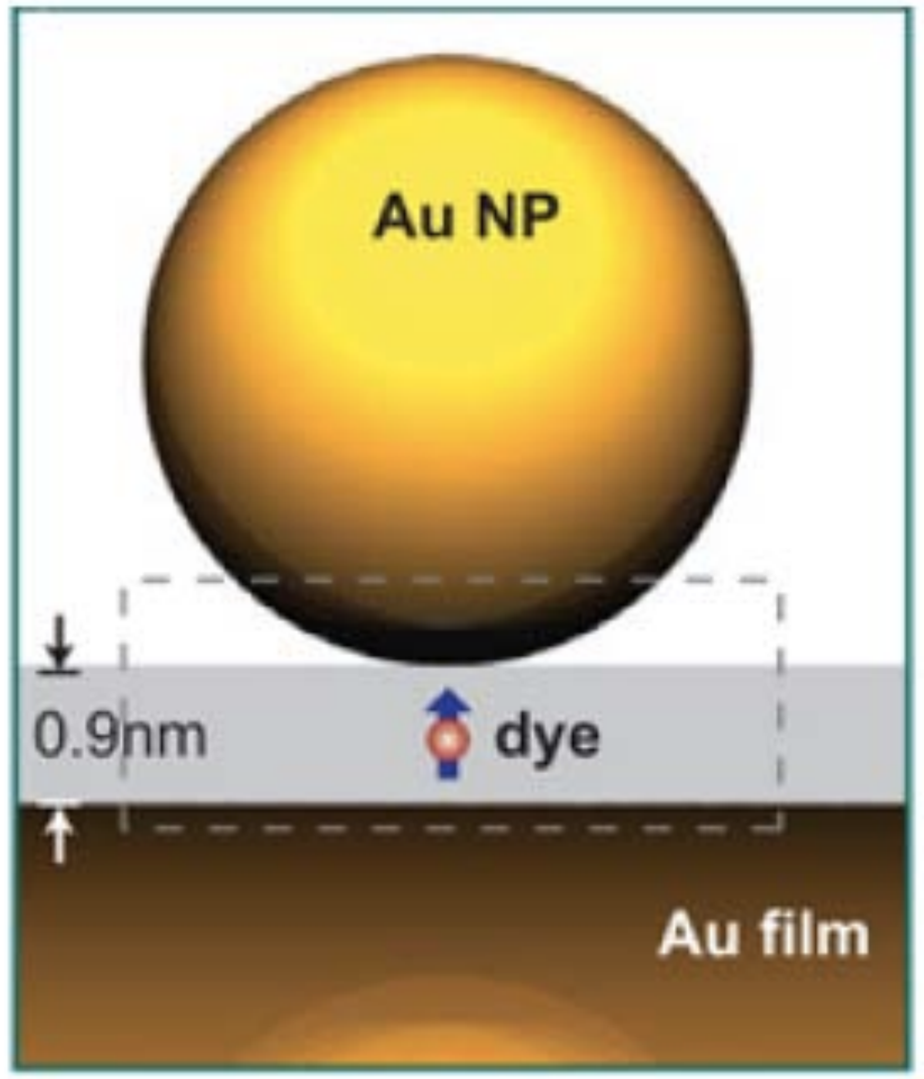
<u>Recent years:</u> Placing atoms and molecules in cavities shown to sometimes dramatically change their properties and chemical reactions: „light-matter (collective) strong coupling".



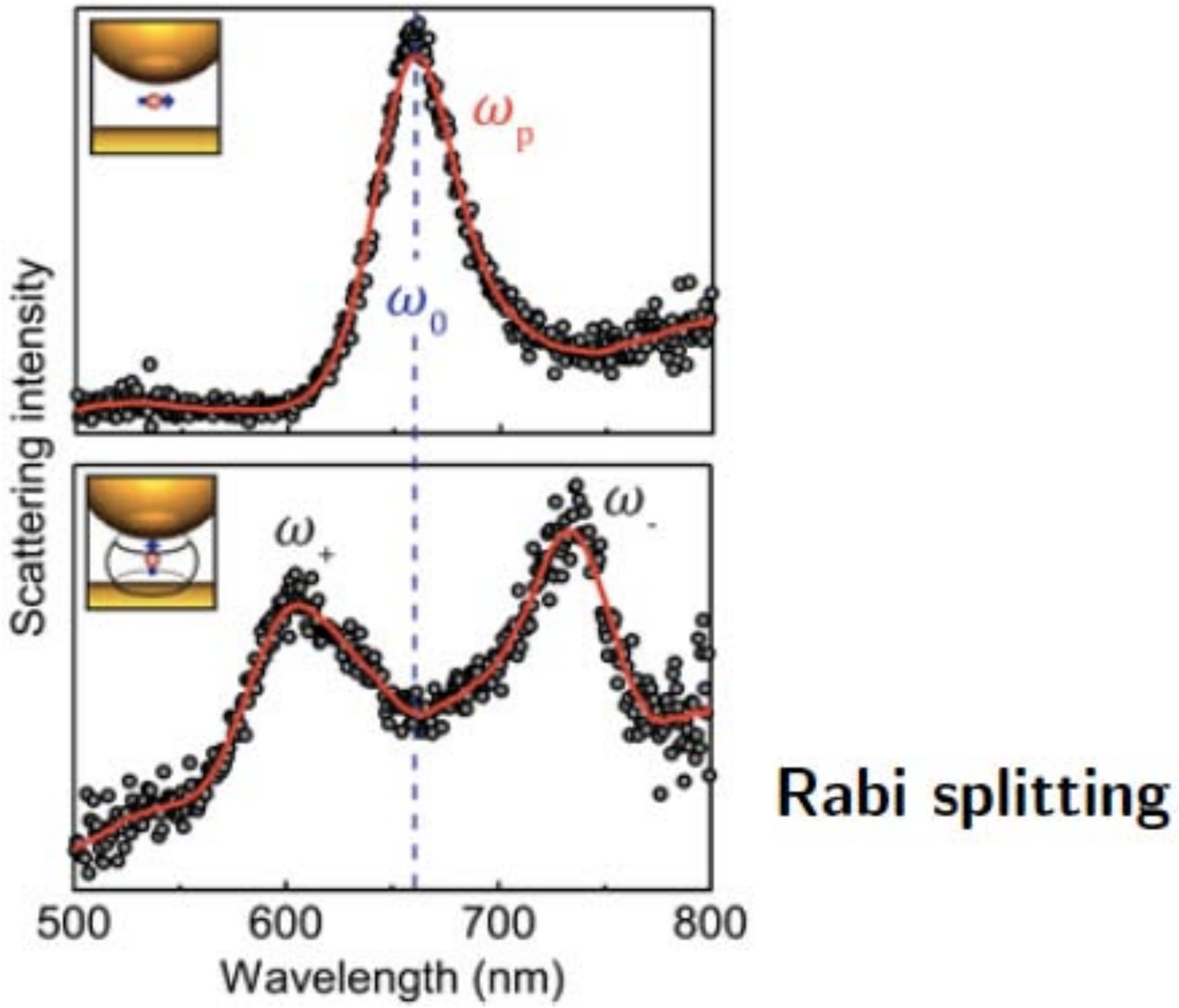
M. Ruggenthaler et al., Nat. Rev. Chem. 2, 0118 (2018)
J. Feist et al., ACS Photonics 5, 205 (2017)
R. F. Ribeiro et al., Chem. Sci. 9, 6325 (2018)
J. Flick et al., Nanophotonics 7, 1479 (2018)
A. F. Kockum et al., Nat. Rev. Phys. 1, 19 (2019)

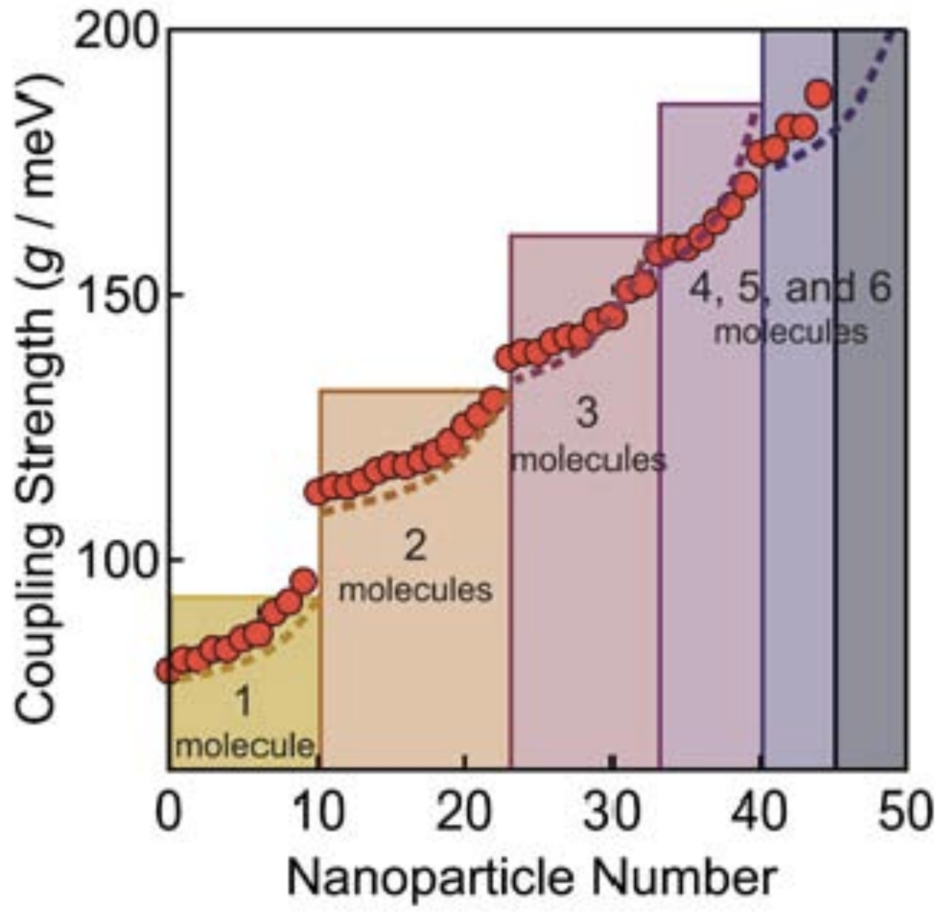
Plasmonic surfaces I





R. Chikkaraddy et al., Nature 535, 127 (2016) – Baumberg group
Also see: K. Santhosh et al., Nature Comm. 7, 11823 (2016) – Gilad Haran group

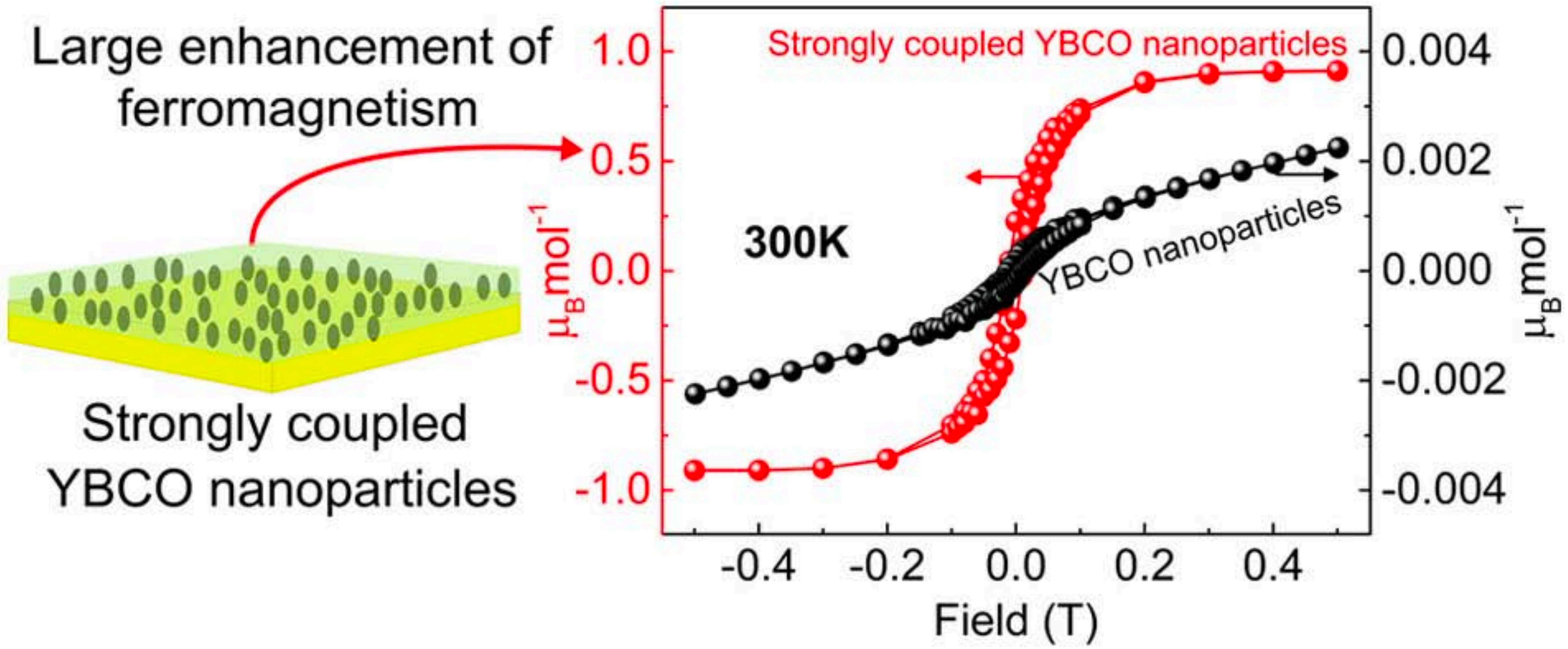




- collective strong light-matter coupling: many atoms interact with the same cavity photon mode
- cavity materials: many atoms interact with same modes
- „Floquet engineering with vacuum fluctuations of light“

Plasmonic surfaces II

Thomas, et int., Ebbesen, Nano Lett. 21, 4365 (2021)

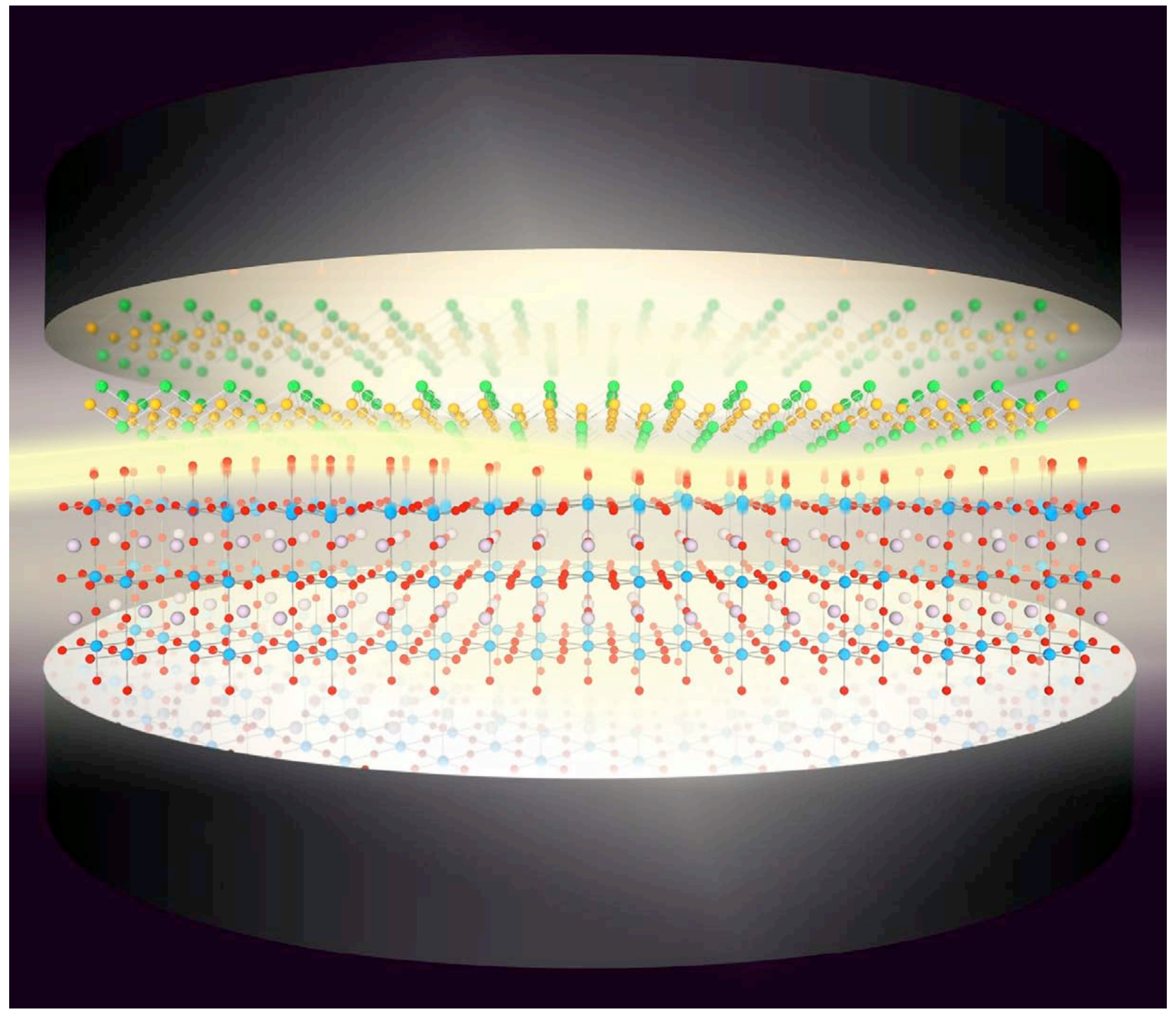


phonon polariton formation influencing electronic properties

Plasmonic surfaces III

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

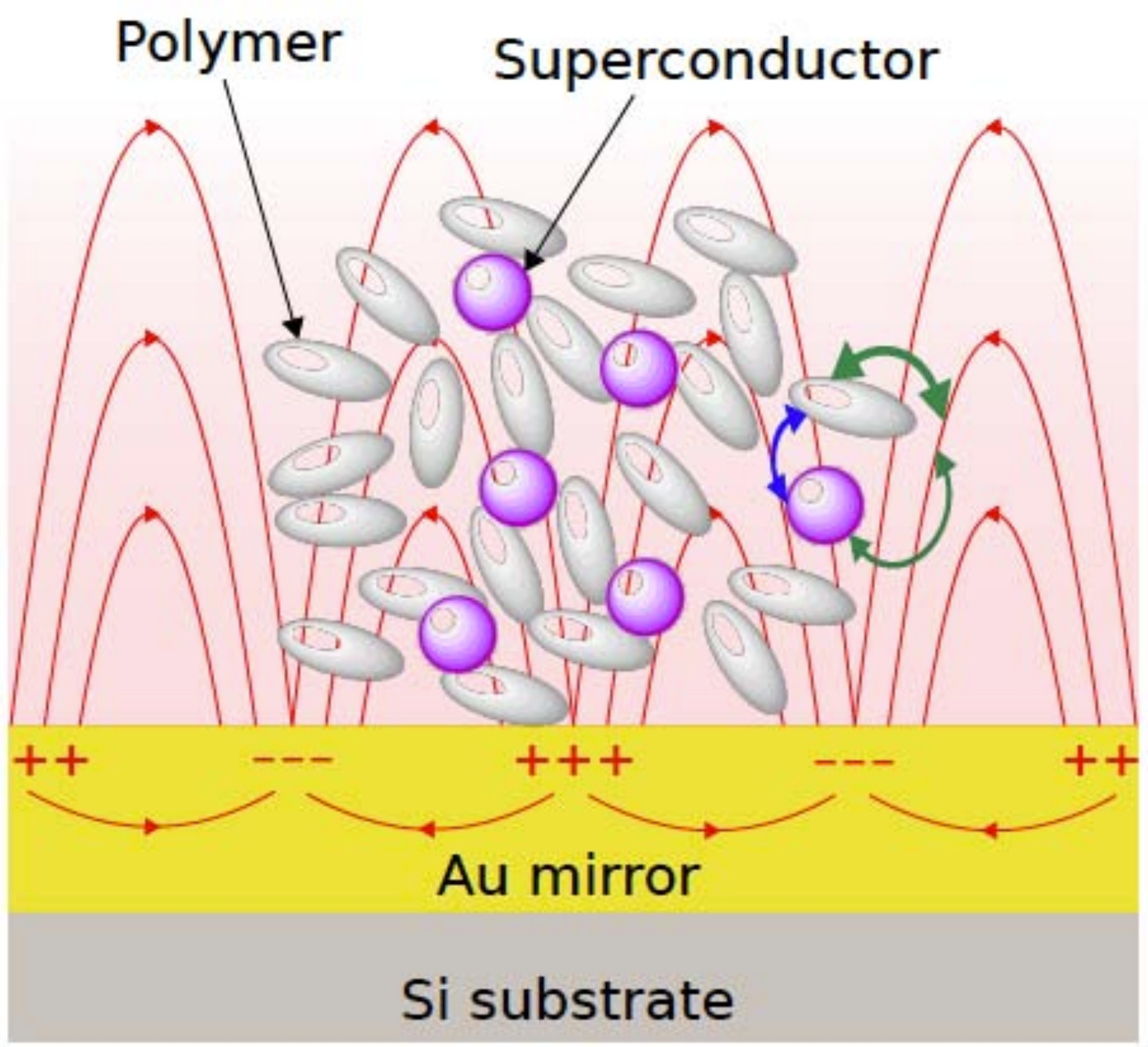
MAS, M. Ruggenthaler, A. Rubio,
Science Advances 4, eaau6969 (2018)



Exploring Superconductivity under Strong Coupling with the Vacuum Electromagnetic Field

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*]

arXiv:1911.01459



Cavity superconductivity?

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,[1] Alexey V. Kavokin,[1,2] and Ivan A. Shelykh[3,4]

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 Cotleț,[1,*] Sina Zeytinoğlu,[1,2] Manfred Sigrist,[2] Eugene Demler,[3] and Ataç Imamoğlu[1]

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

Hagenmüller et al., 1801.09876

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

M. A. Sentef,[1,*] M. Ruggenthaler,[1] and A. Rubio[1,2,3]

1802.09437

Cavity-mediated electron-photon superconductivity *1804.07142*

Frank Schlawin[1], Andrea Cavalleri[1,2] and Dieter Jaksch[1]

Superradiant Quantum Materials *1804.08534*

Giacomo Mazza[1,2,*] and Antoine Georges[2,3,1,4]

Cavity Quantum Eliashberg Enhancement of Superconductivity

Jonathan B. Curtis,[1,2,*] Zachary M. Raines,[1,2] Andrew A. Allocca,[1,2] Mohammad Hafezi,[1] and Victor M. Galitski[1,2] *1805.01482*

Manipulating quantum materials with quantum light *1806.06752*

Martin Kiffner[1,2], Jonathan Coulthard[2], Frank Schlawin[2], Arzhang Ardavan[2], and Dieter Jaksch[2,1]

Cavity superconductor-polaritons *1807.06601*

Andrew A. Allocca,* Zachary M. Raines, Jonathan B. Curtis, and Victor M. Galitski

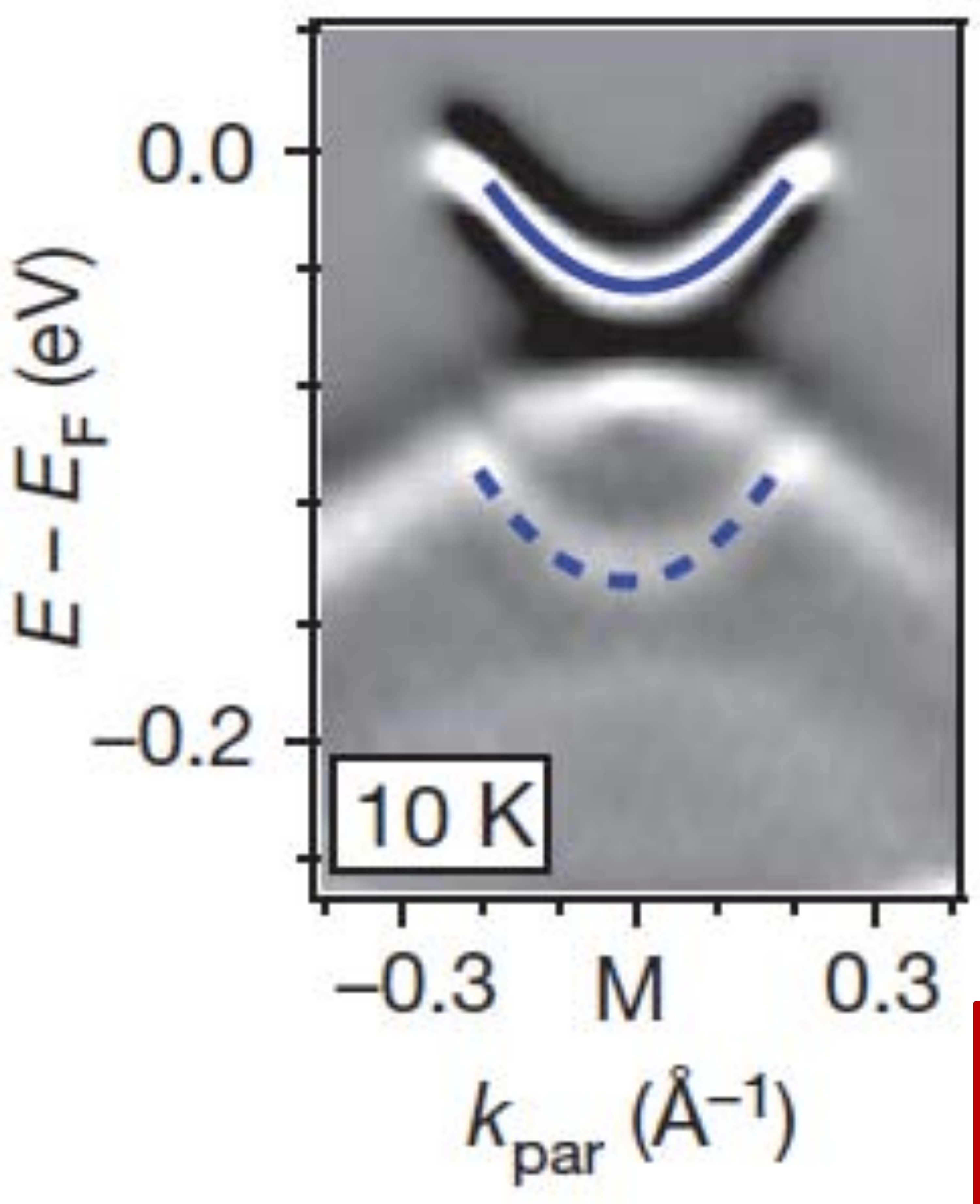
Ab-initio Exciton-polaritons:
Cavity control of Dark Excitons in two dimensional Materials

Simone Latini,[1,*] Enrico Ronca,[1,†] Umberto De Giovannini,[1,2,‡] Hannes Hübener,[1,§] and Angel Rubio[1,3,¶]

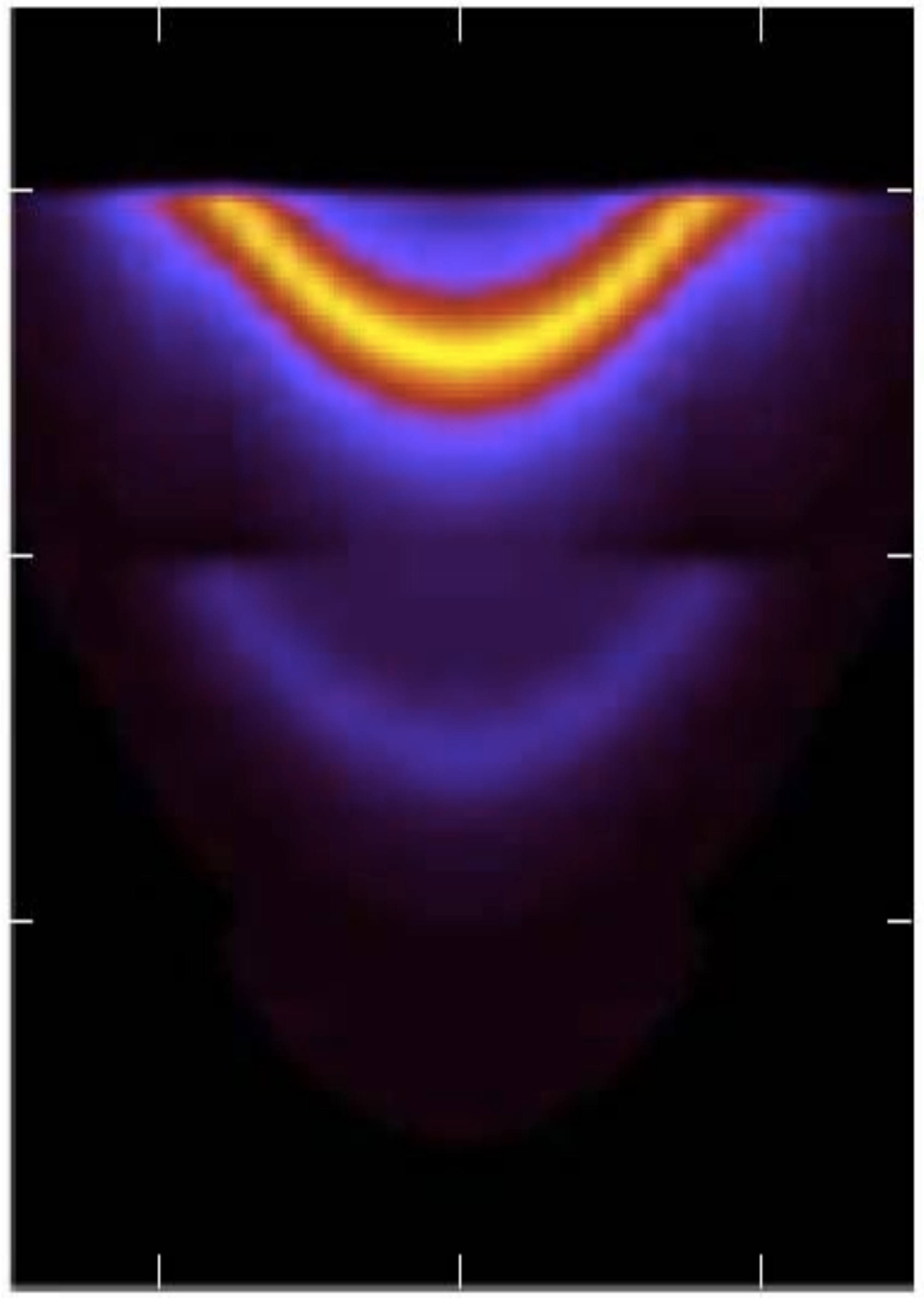
1810.02672

monolayer FeSe/STO: ARPES

experiment



theory



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

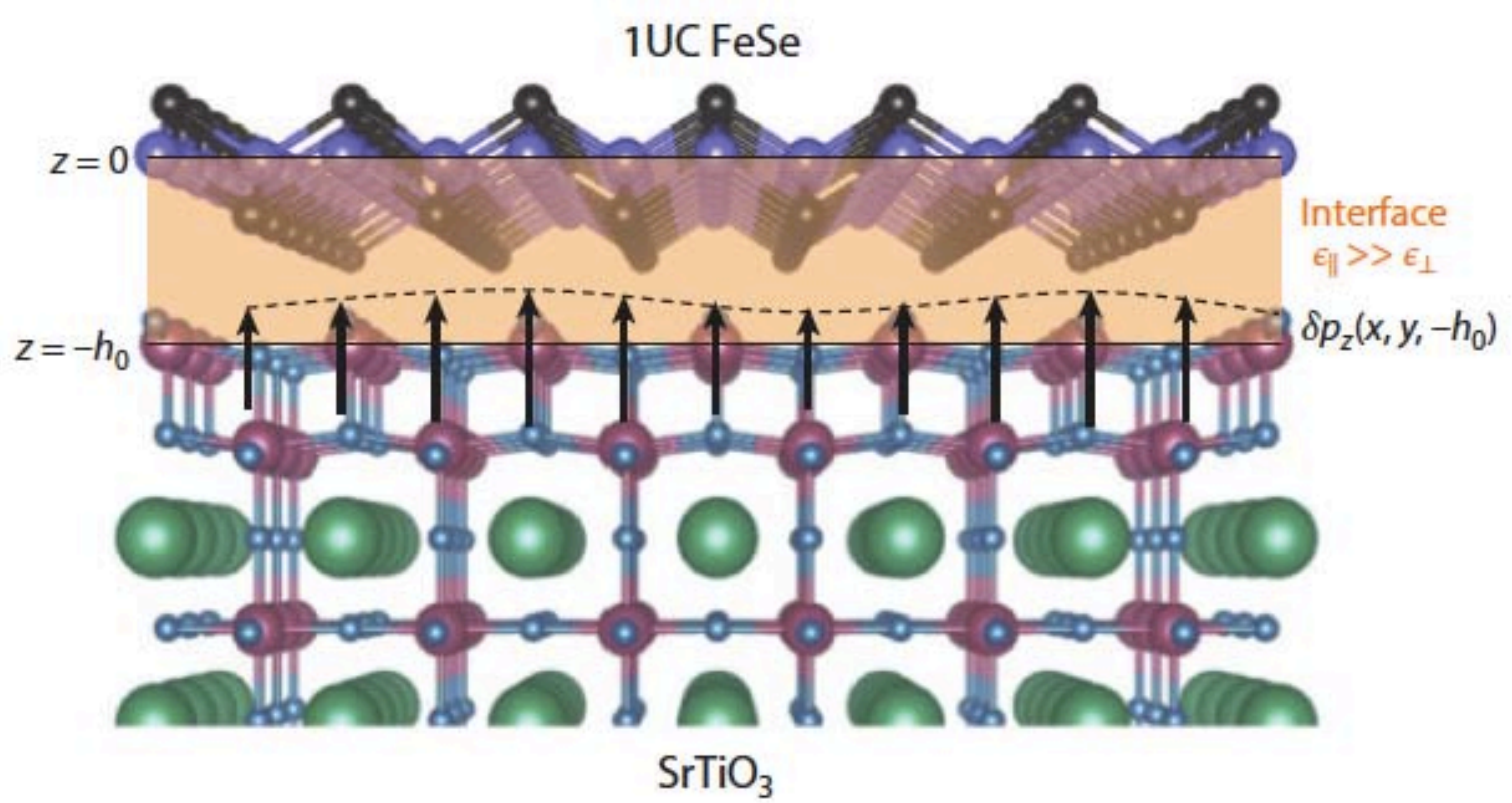
Lee et al., Nature 515, 245 (2014)

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)*

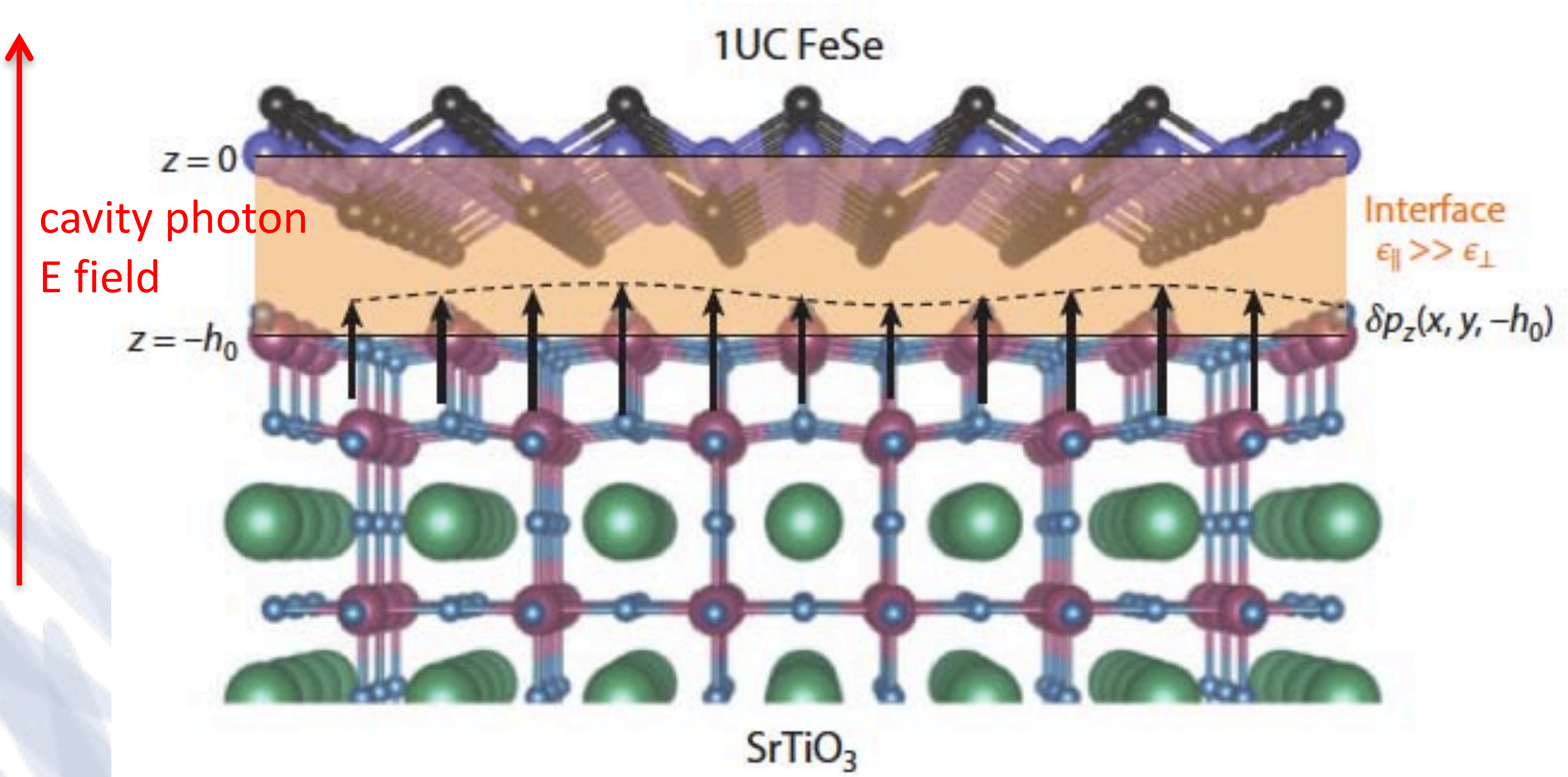
$$q_0^{-1} = h_0\sqrt{\epsilon_{\|}/\epsilon_{\perp}} \quad \epsilon_{\|}/\epsilon_{\perp} \approx 100$$



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)

Model and Method

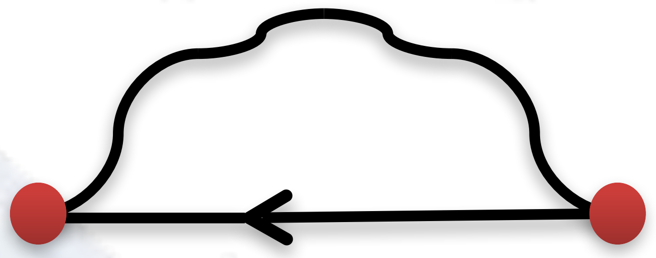
electrons — el-polariton coupling — polaritons

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

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

G-self-consistent Migdal-Eliashberg diagram

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

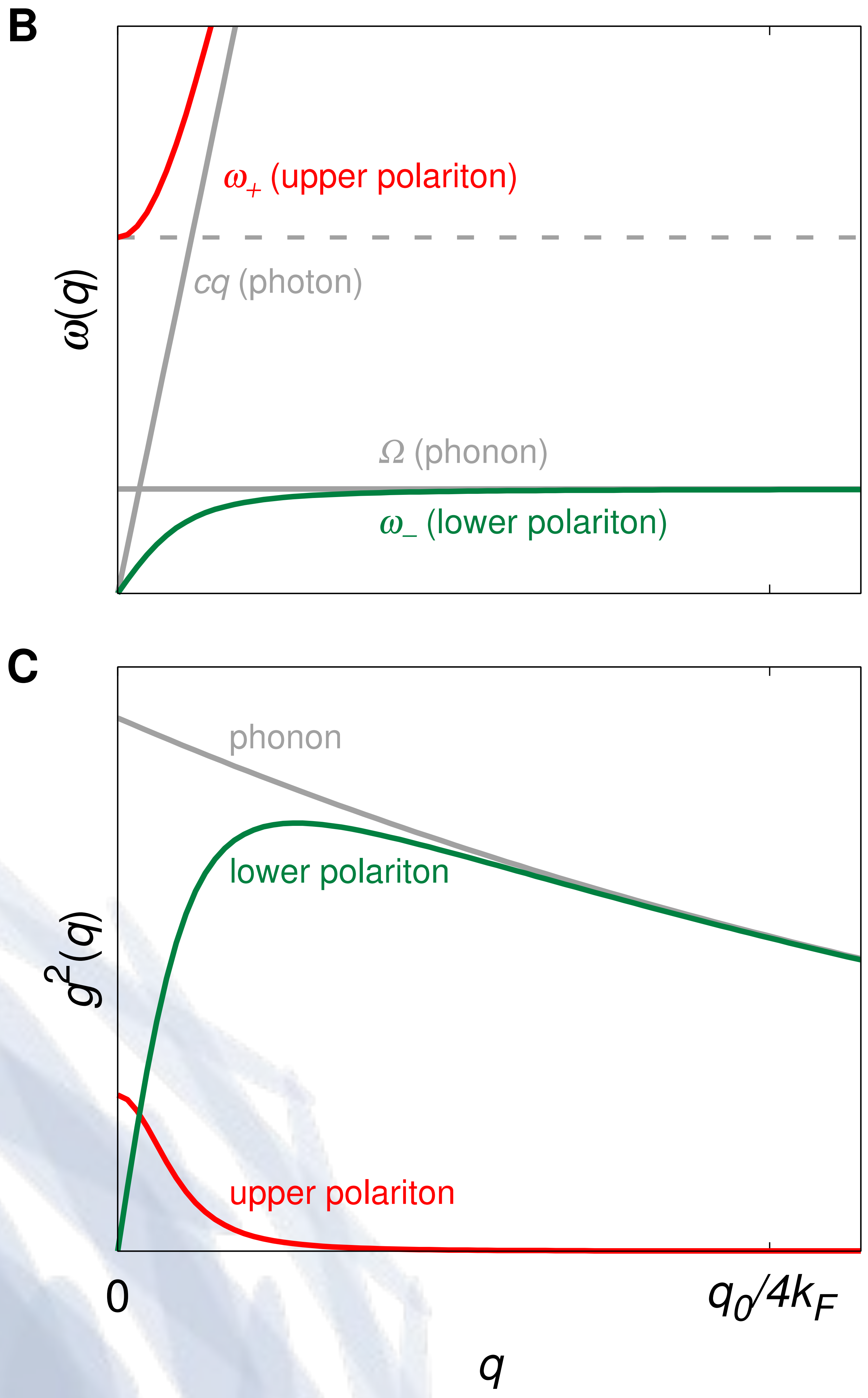


$$\hat{\Sigma}(\vec{k},i\omega_n)=i\omega_n[1-Z(\vec{k},i\omega_n)]\hat{\tau}_0+\chi(\vec{k},i\omega_n)\hat{\tau}_3+\phi(\vec{k},i\omega_n)\hat{\tau}_1$$

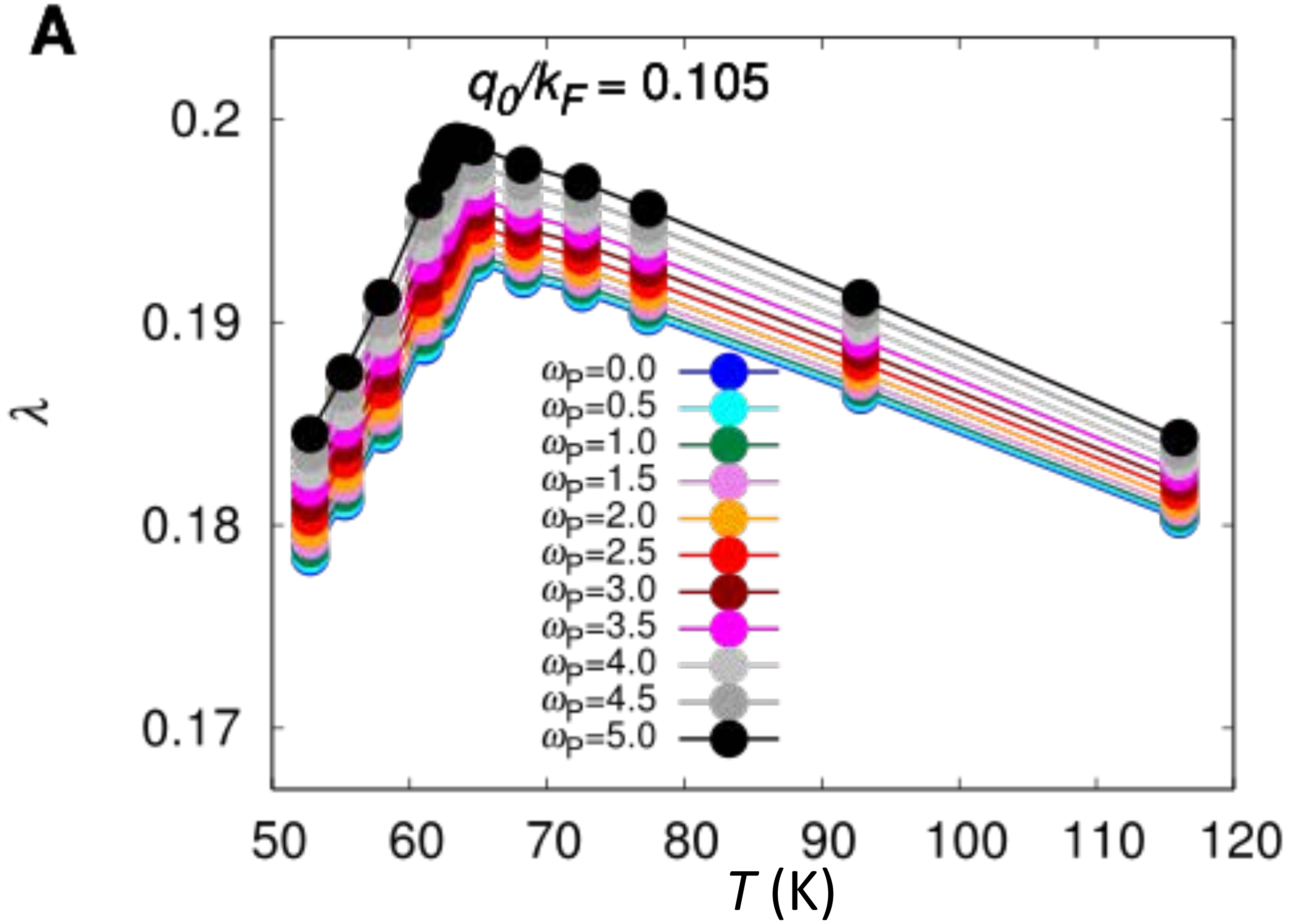
$$\lambda\equiv Z(\vec{k}_F,i\pi/\beta)-1$$

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

Cavity materials: Phonon polaritons

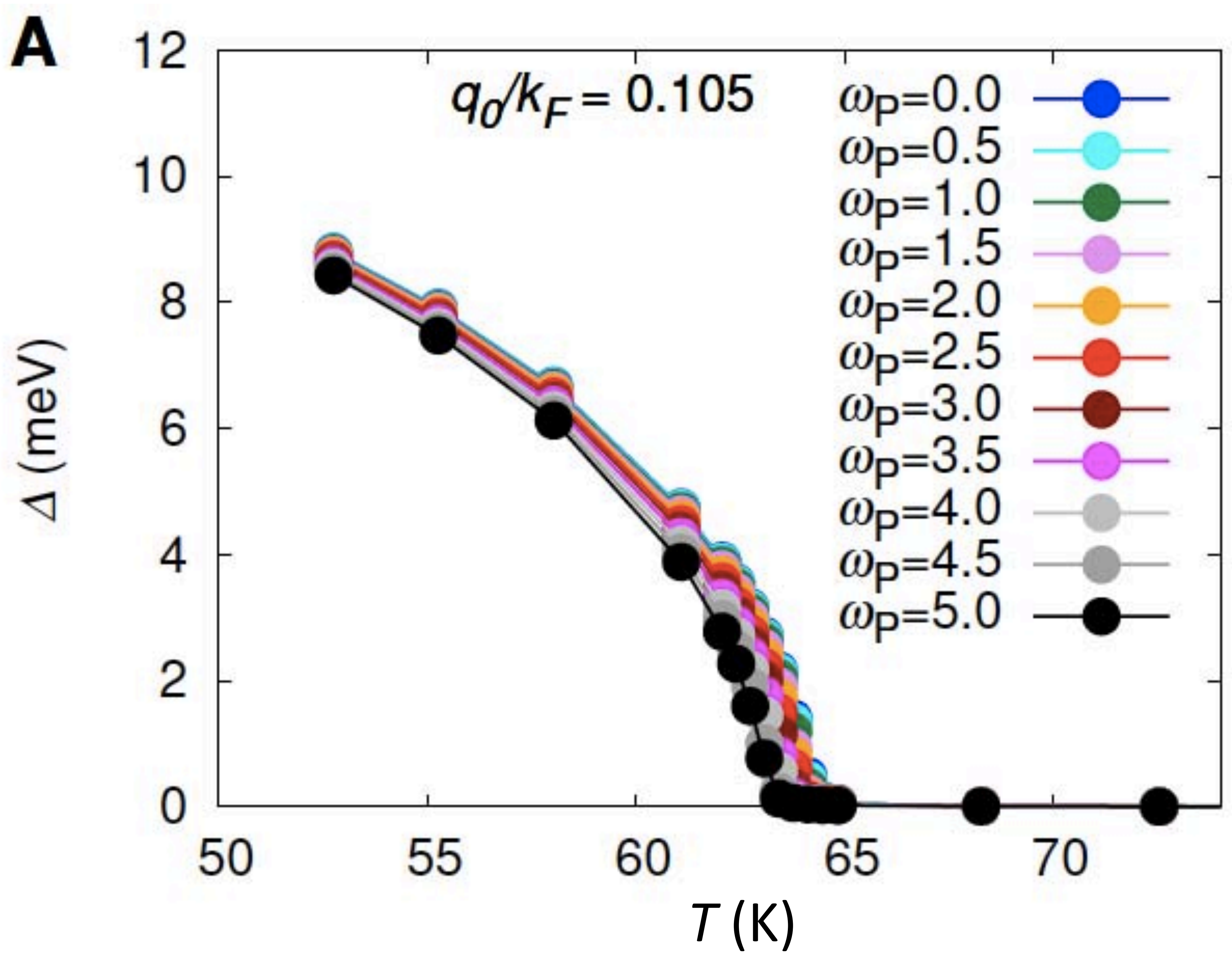


Migdal-Eliashberg theory



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.

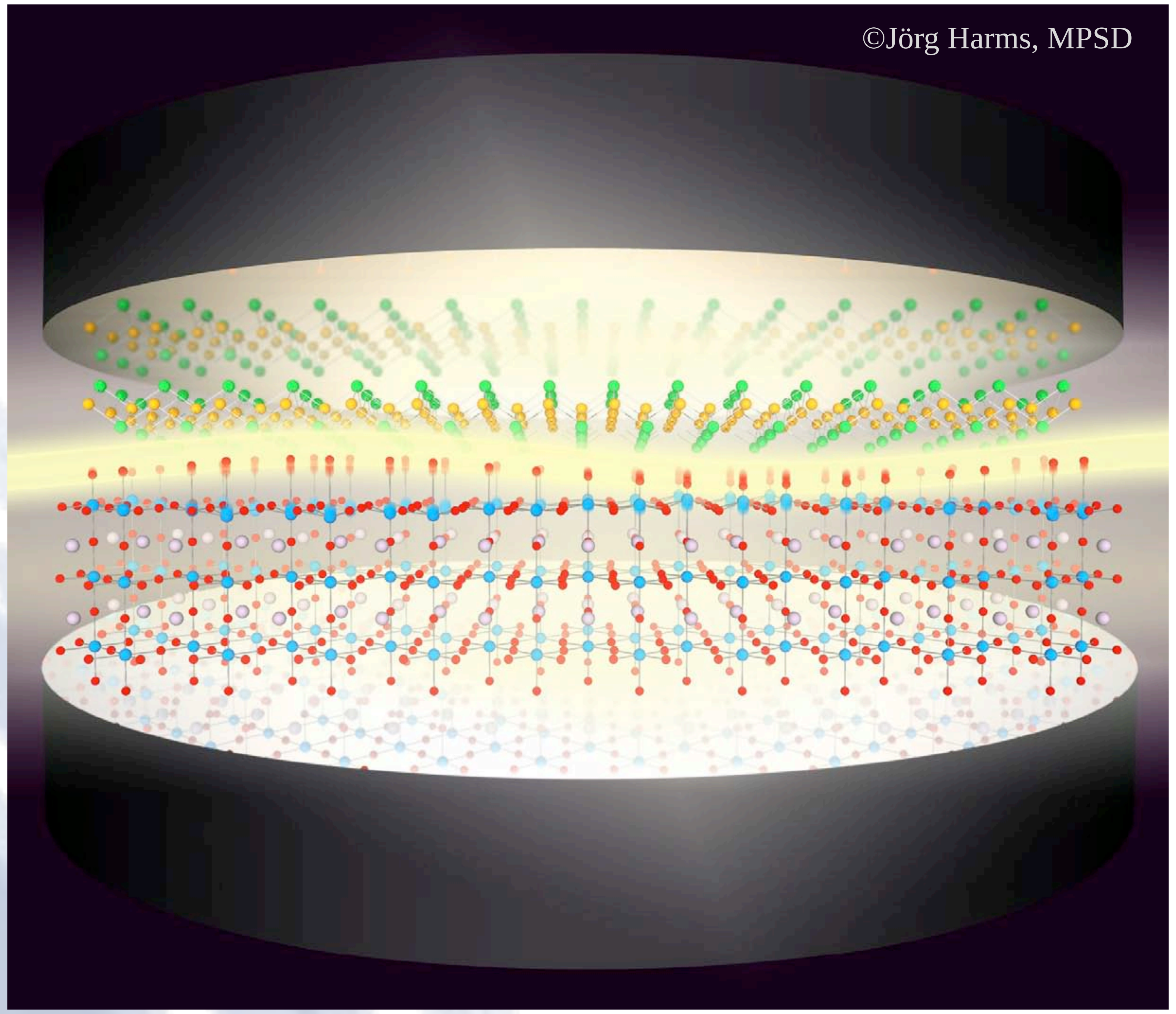
$$T_{C,\mathrm{BCS}} \approx 1.13\Omega\exp(-\tfrac{1}{\lambda})$$

q-independent scattering

Cavity-modified superconductivity

- 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)

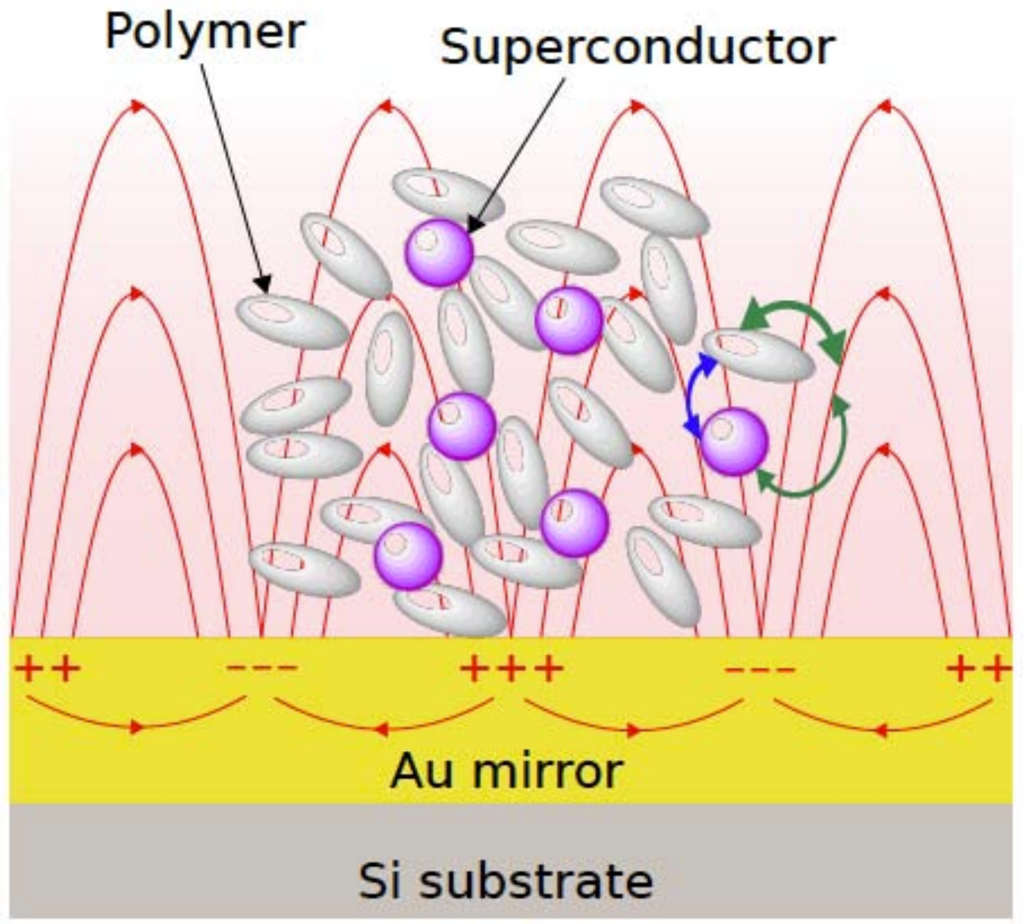


The jury is still out …

Exploring Superconductivity under Strong Coupling with the Vacuum Electromagnetic Field

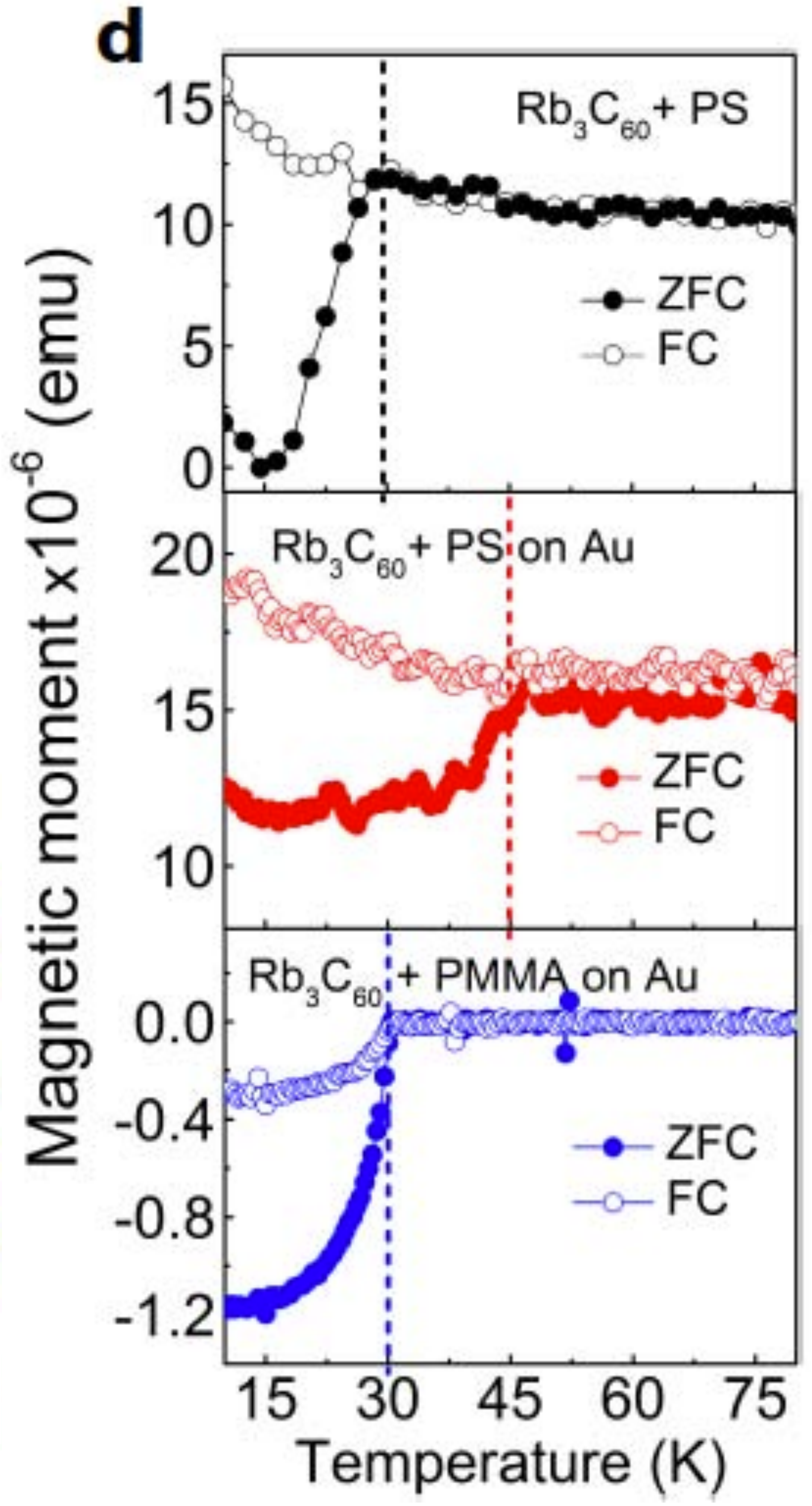
arXiv:1911.01459

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*]

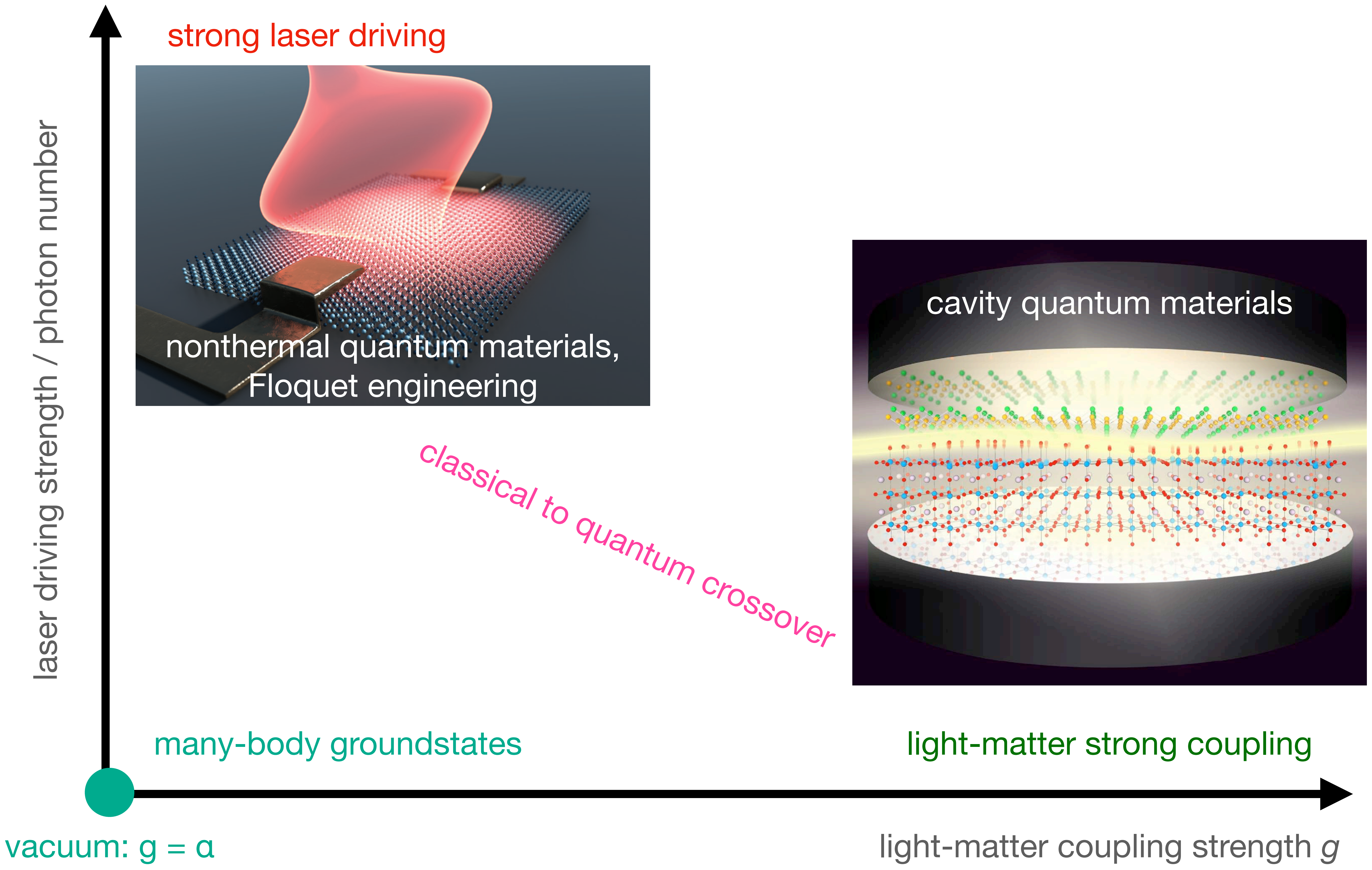


... consistent with enhanced electron-phonon coupling due to polariton formation and mode softening

By placing the superconductor-surface plasmon system in a SQUID magnetometer, we find that the superconducting transition temperatures (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.

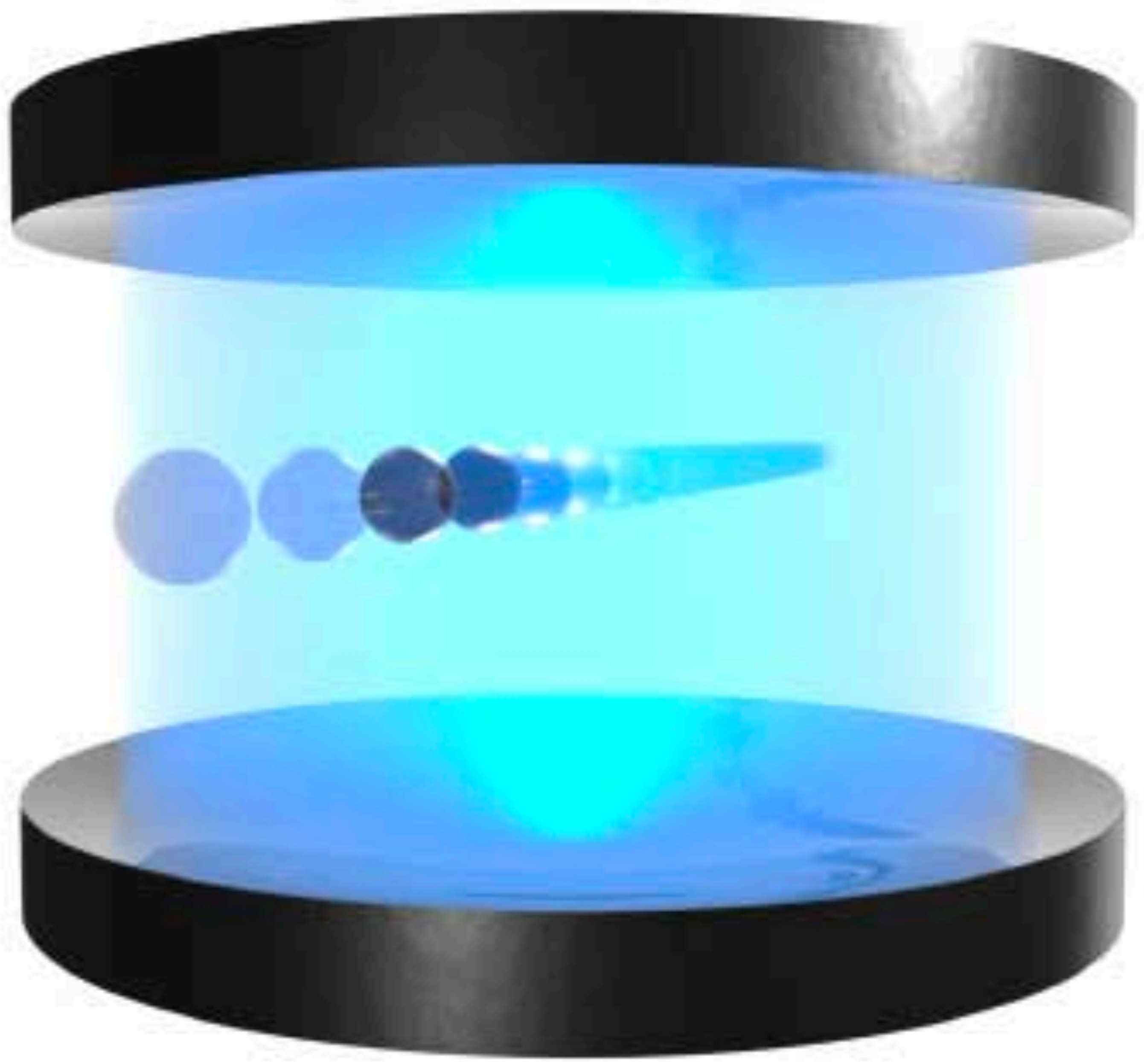


Can we employ light-matter interactions to change materials properties?

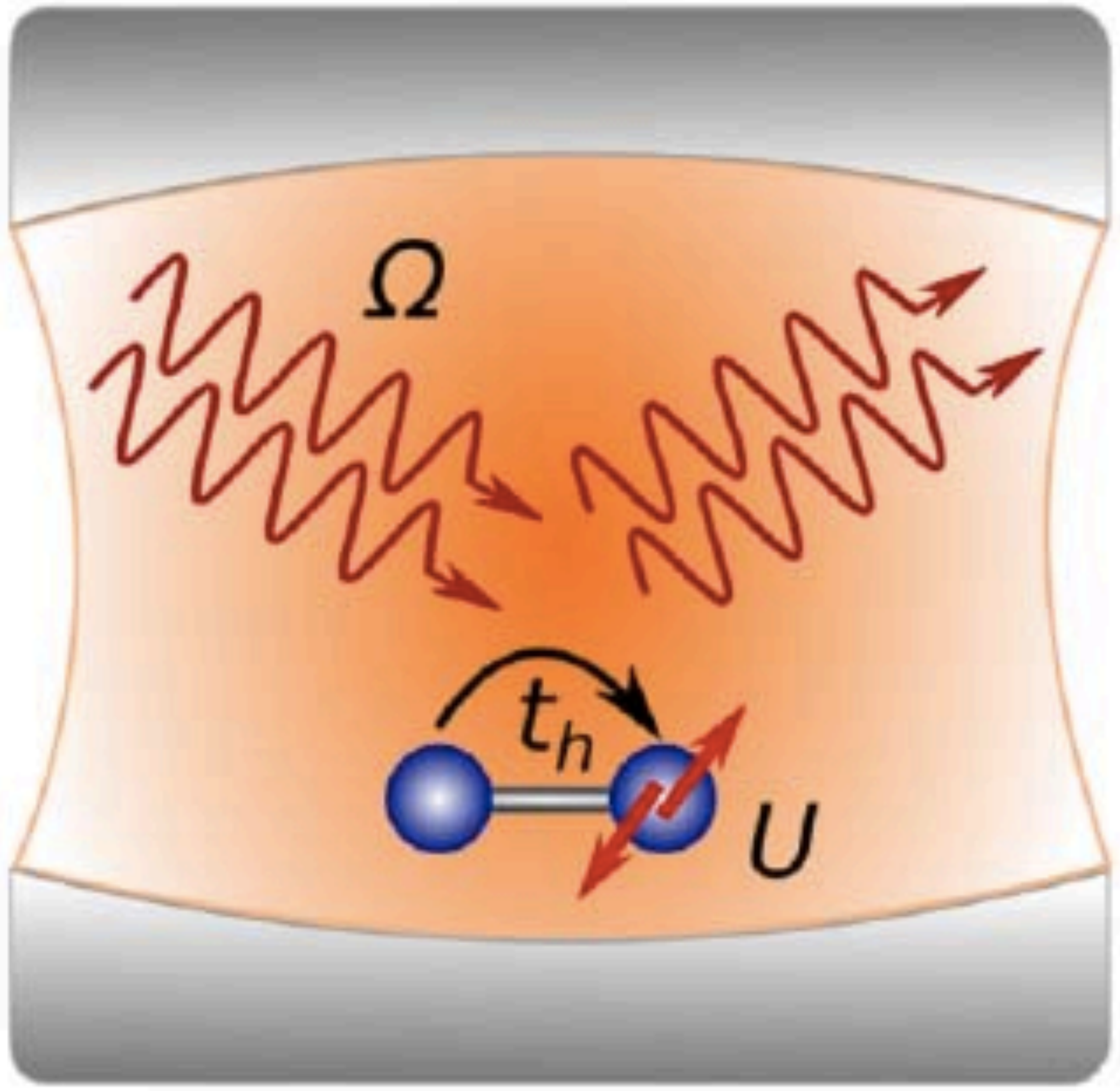


Quantum to classical crossover

Quantum Floquet engineering with an exactly solvable tight-binding chain in a cavity,
C. J. Eckhardt, G. Passetti, et al.,
Communications Physics 5, 122 (2022)

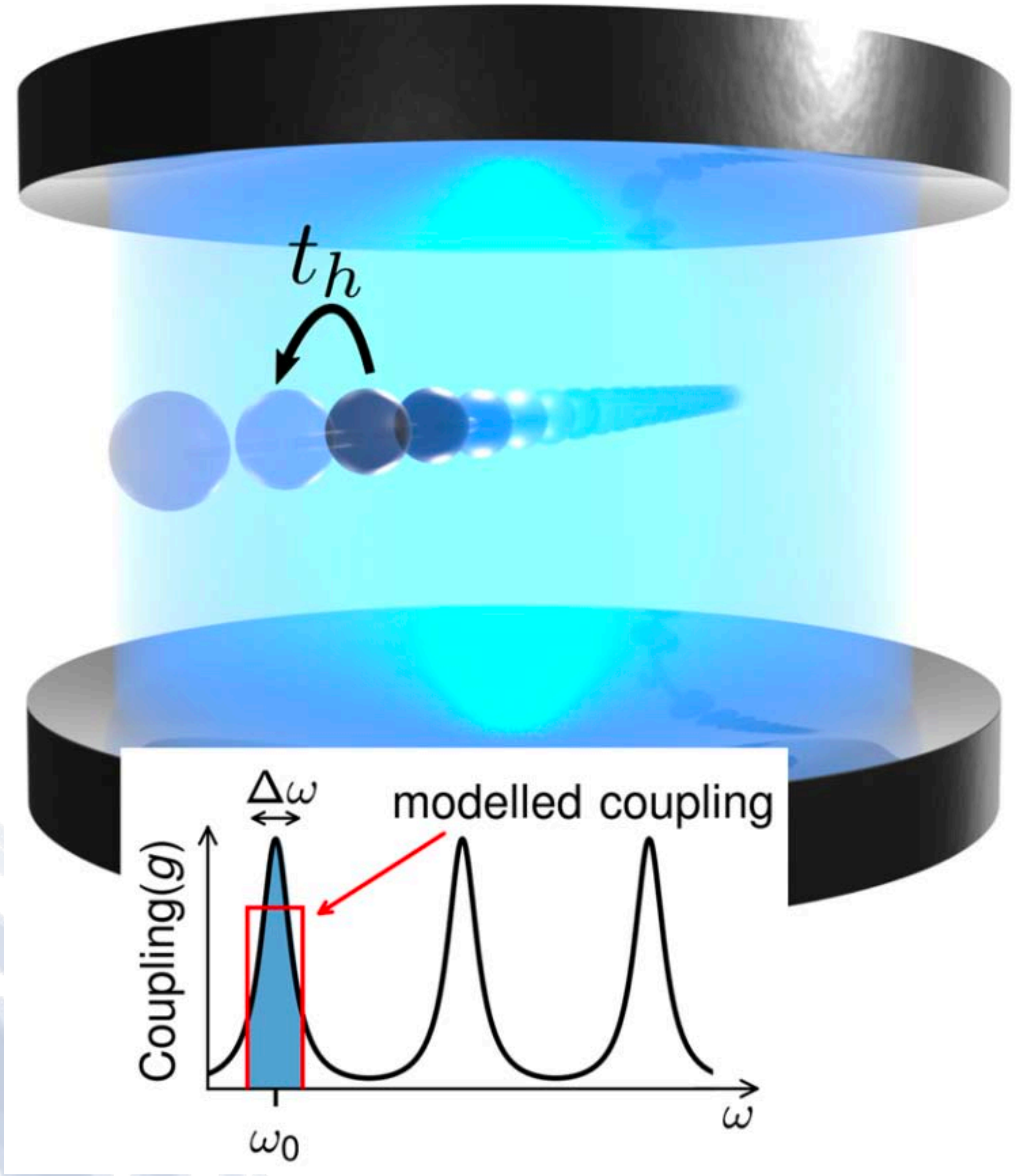


Quantum to classical crossover of Floquet engineering in correlated quantum systems,
MAS, J. Li, F. Künzel, M. Eckstein,
PRResearch 2, 033033 (2020)



Quantum chain in cavity

Quantum Floquet engineering with an exactly solvable tight-binding chain in a cavity,
C. J. Eckhardt, G. Passetti, et al.,
Communications Physics 5, 122 (2022)



$$H = \omega_0\left(a^\dagger a + \frac{1}{2}\right) - \sum_{j=1}^{L}\left[t_h e^{-i\frac{g}{\sqrt{L}}(a^\dagger + a)}\, c^\dagger_{j+1}c_j + \text{h.c.}\right]$$

$$H = \cos\left(\frac{g}{\sqrt{L}}(a+a^\dagger)\right)\mathcal{T} + \sin\left(\frac{g}{\sqrt{L}}(a+a^\dagger)\right)\mathcal{J} + \omega_0\left(a^\dagger a + \frac{1}{2}\right)$$

kinetic energy

$$\mathcal{T} := \sum_k -2t_h \cos(k)\, c^\dagger_k c_k =: \sum_k \varepsilon_k\, c^\dagger_k c_k$$

current

$$\mathcal{J} := \sum_k 2t_h \sin(k)\, c^\dagger_k c_k =: \sum_k v_k\, c^\dagger_k c_k,$$

$$\rho_k = c^\dagger_k c_k \qquad [\rho_k, H] = 0 \quad \text{for all} \quad k \in \text{BZ}$$

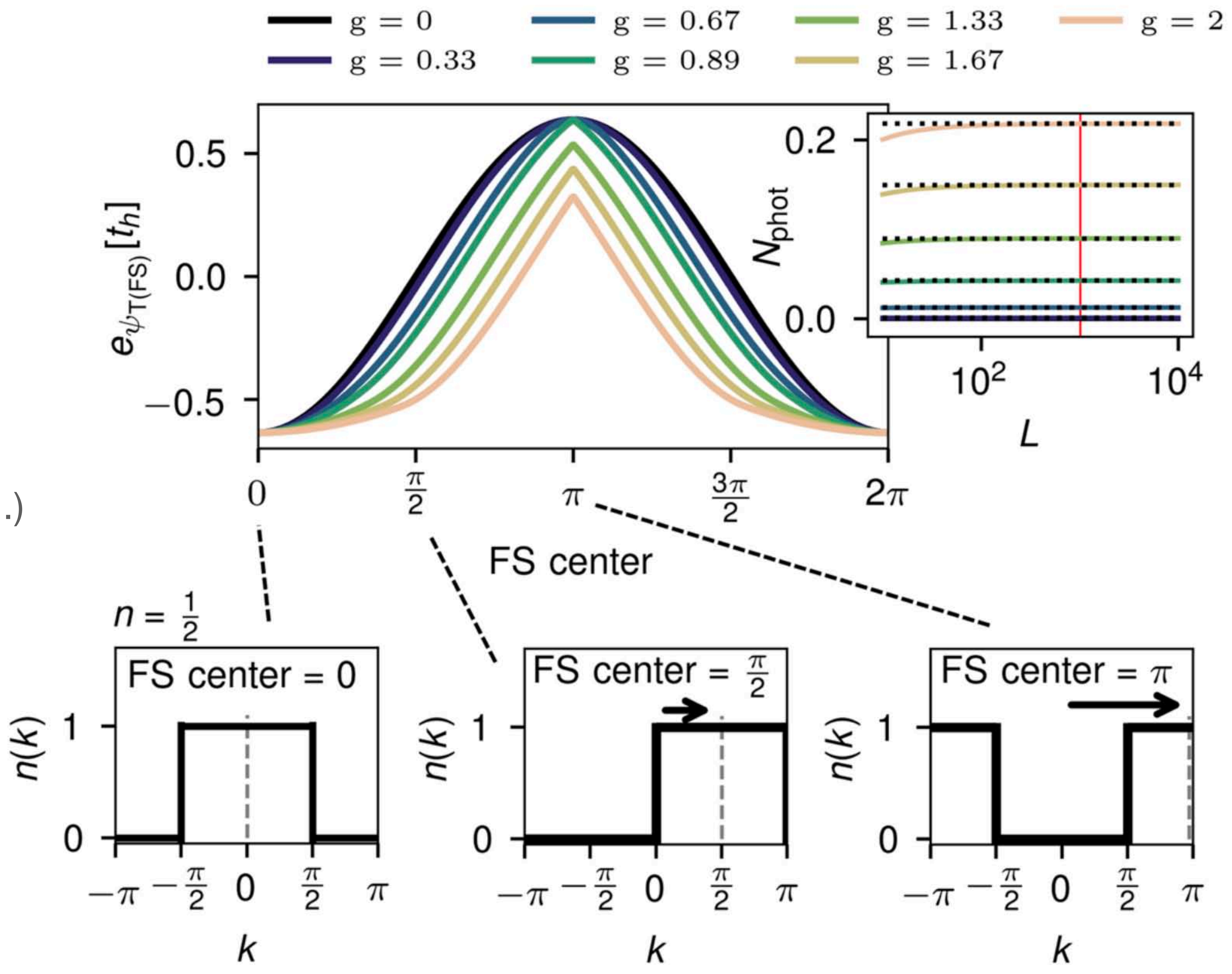
macroscopic # of constants of motion!

(also cf. Li, Golez, Mazza, Millis, Georges, Eckstein PRB 2020)

Quantum chain in cavity

Variational search for exact
cavity-chain coupled groundstate:

Fermi sea around k=0
electronic wave function unchanged
(but we will see that excitation spectrum is different…)



Quantum chain in cavity

Groundstate

$$H = \tilde{\omega}(a^\dagger a + \frac{1}{2}) + T + \frac{1}{24}\frac{g^4\tilde{\omega}^2}{L^2\omega_0^2}(a^\dagger + a)^4 T + \dots \quad ; \quad \tilde{\omega} = \omega_0\sqrt{1 - 2\frac{g^2}{\omega_0 L}T}$$

$$|\Phi\rangle = e^{-S^{sq}}|0\rangle \; ; \quad S^{sq} = \frac{1}{2}\zeta((a^\dagger)^2 - a^2)$$

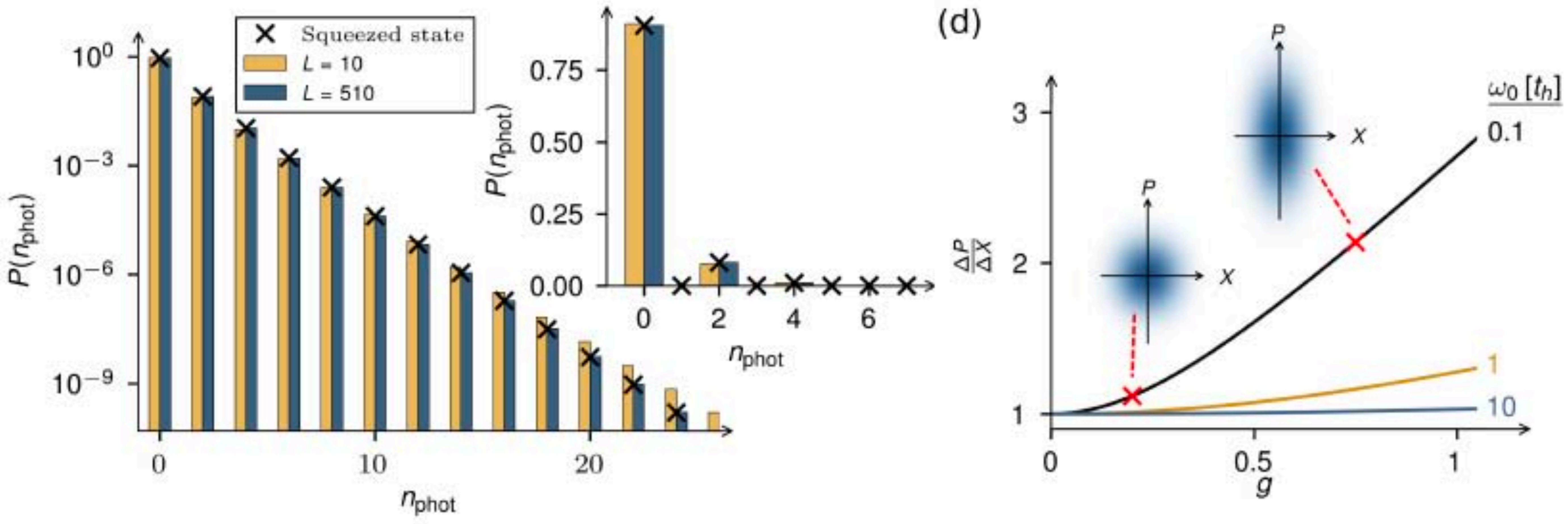
$$\zeta = \frac{1}{4}\ln(1 - 2\frac{g^2}{L\omega_0}T)$$

Squeezed state of cavity mode
Degree of squeezing controlled through strength of light-matter coupling

Prediciton of Vacuum squeezed state (different model):
Ciuti, C., Bastard, G., Carusotto, I. Q, PRB (2005)

Direct sampling of vacuum fluctuations
Benea-Chelmus, I.-C., Settembrini, F. F., Scalari, G., Faist, J., Nature (2019)

C. Riek, D. V. Seletskiy, A. S. Moskalenko, J. F. Schmidt, P. Krauspe, S. Eckart, S. Eggert, G. Burkard, A. Leitenstorfer, Science (2015)



Quantum chain in cavity

Quantum Floquet engineering with an exactly solvable tight-binding chain in a cavity,
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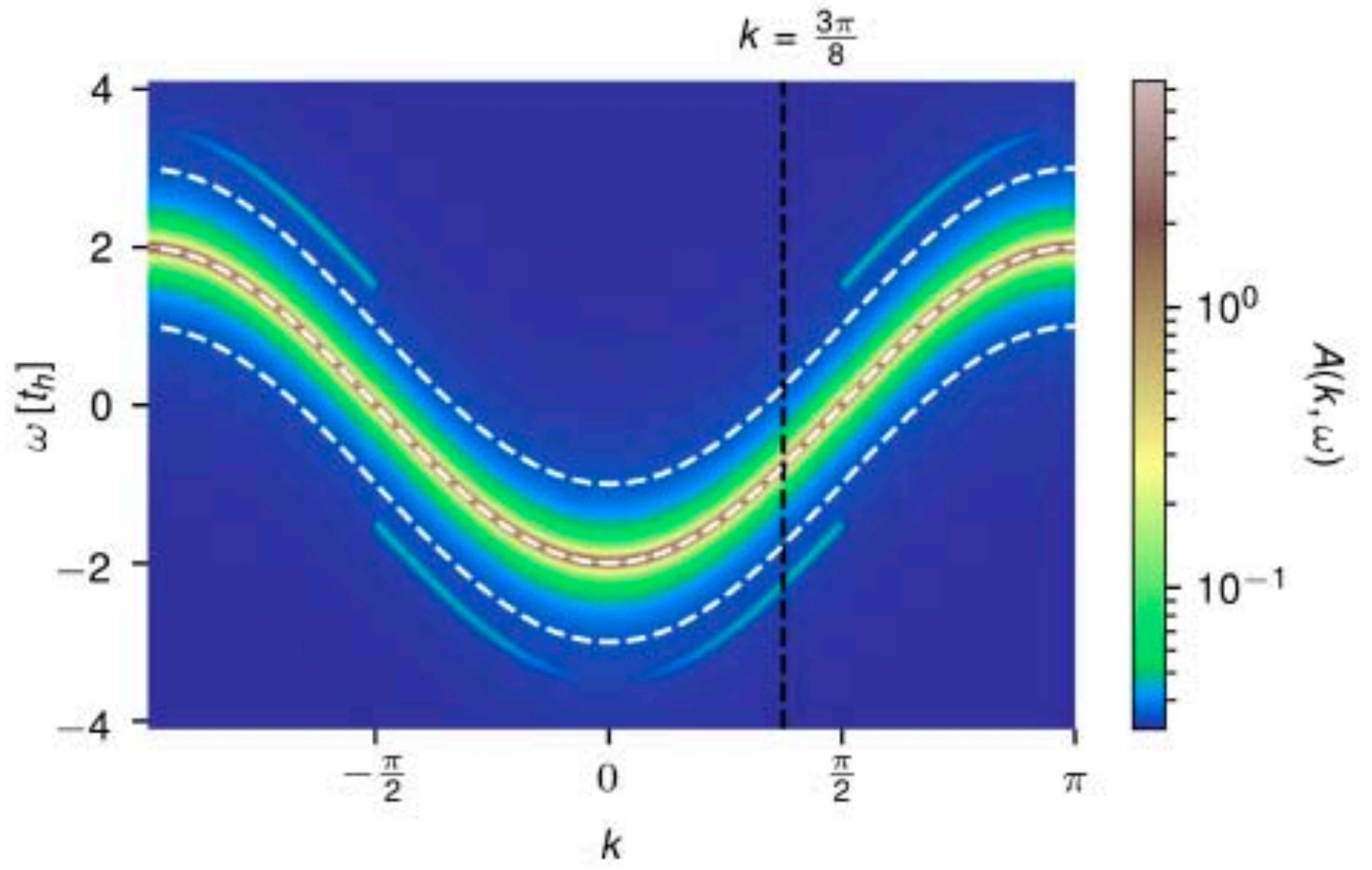
Communications Physics 5, 122 (2022)



Electronic spectral function in cavity is changed
(despite GS being unshifted Fermi sea)
-> photon shakeoff states (Franck-Condon like)
-> precursor of „Floquet sidebands“?

Q: Which other models do we know where the groundstate (of a subsystem) is unchanged but excitation spectrum is changed? Is it interesting?

$$A(k,\omega) = 2\pi\delta(\omega-\varepsilon_k) + \mathcal{O}(\frac{1}{L}) = 2\pi n_k e^{-\lambda}\sum_\ell \frac{\lambda^\ell}{\ell!}\delta(\omega-\varepsilon_k(1-\frac{g^2}{2L}\frac{\omega_0}{\tilde{\omega}})-\Sigma_k+\tilde{\omega}\ell)$$
$$+ 2\pi(1-n_k)e^{-\lambda}\sum_\ell \frac{\lambda^\ell}{\ell!}\delta(\omega-\varepsilon_k(1-\frac{g^2}{2L}\frac{\omega_0}{\tilde{\omega}})-\Sigma_k+\tilde{\omega}\ell)$$

generalization of textbook result (Poisson dist., „Franck-Condon ladder“, cf. Mahan book)

Quantum chain in cavity

Quantum to Classical Crossover

Prepare the cavity in a coherent state

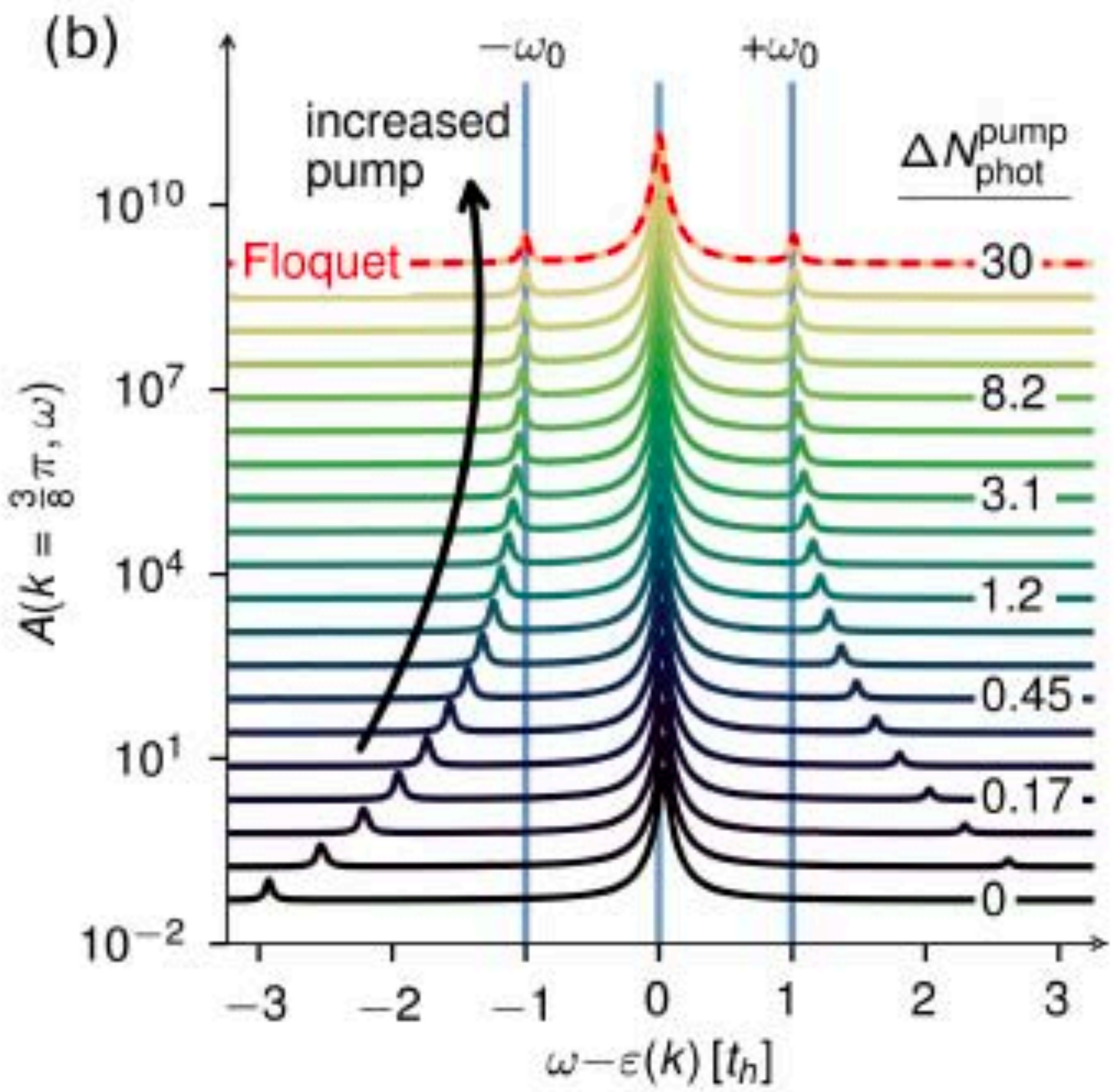
$$|\psi\rangle = |e^{-}_{GS}\rangle \otimes |\mathrm{coh}(N)\rangle$$

TD Limit:

$$L \to \infty\,,\; A \sim \frac{1}{\sqrt{L}}$$

Quantum to Classical:

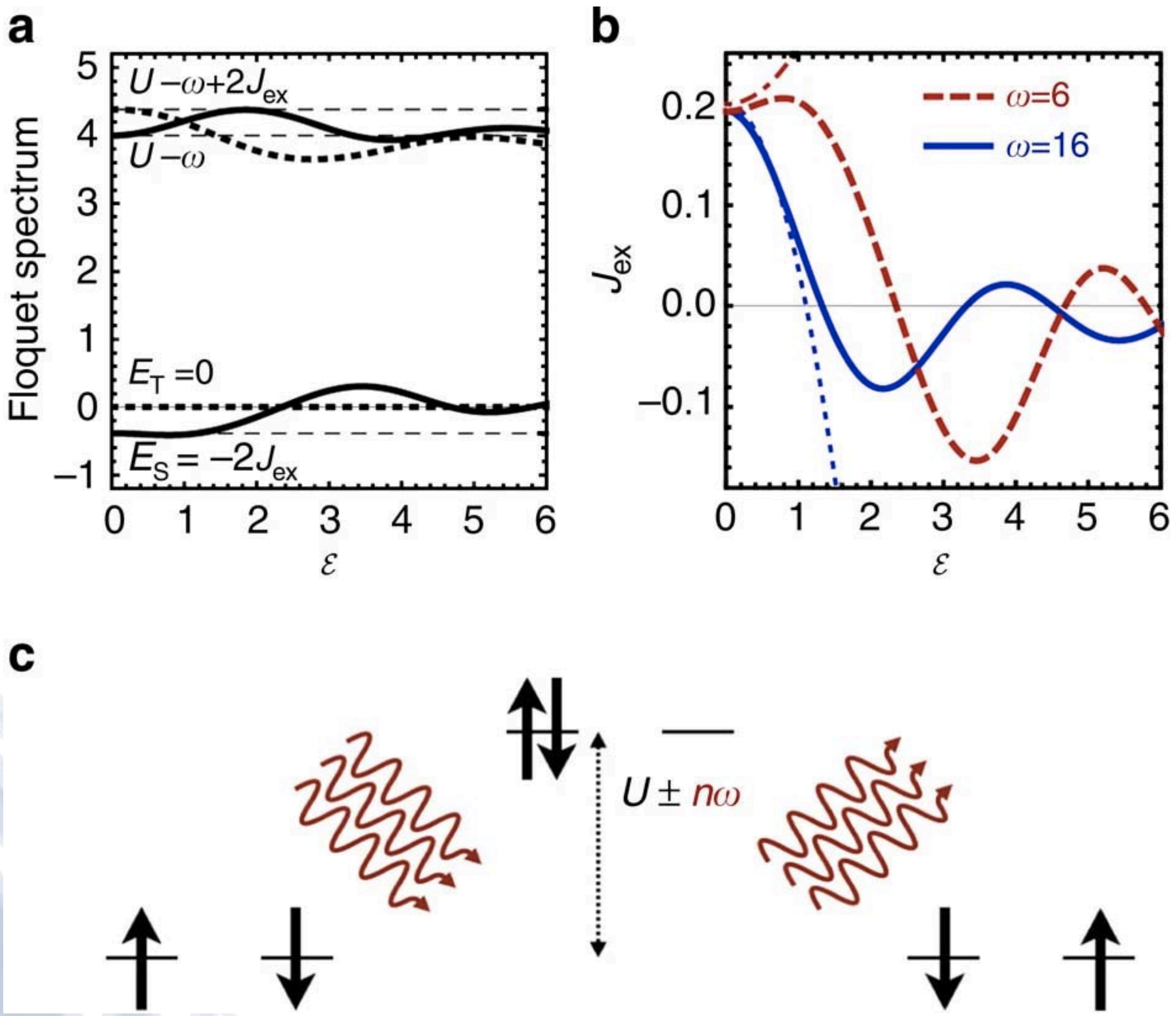
$$N_{pt} \to \infty\,,\; A \sim \frac{1}{\sqrt{N_{pt}}}$$



Floquet engineering of quantum materials: interactions?

Floquet engineering of spin exchange

Mentink, Balzer, and Eckstein, Nat. Commun. 6, 6708 (2015)



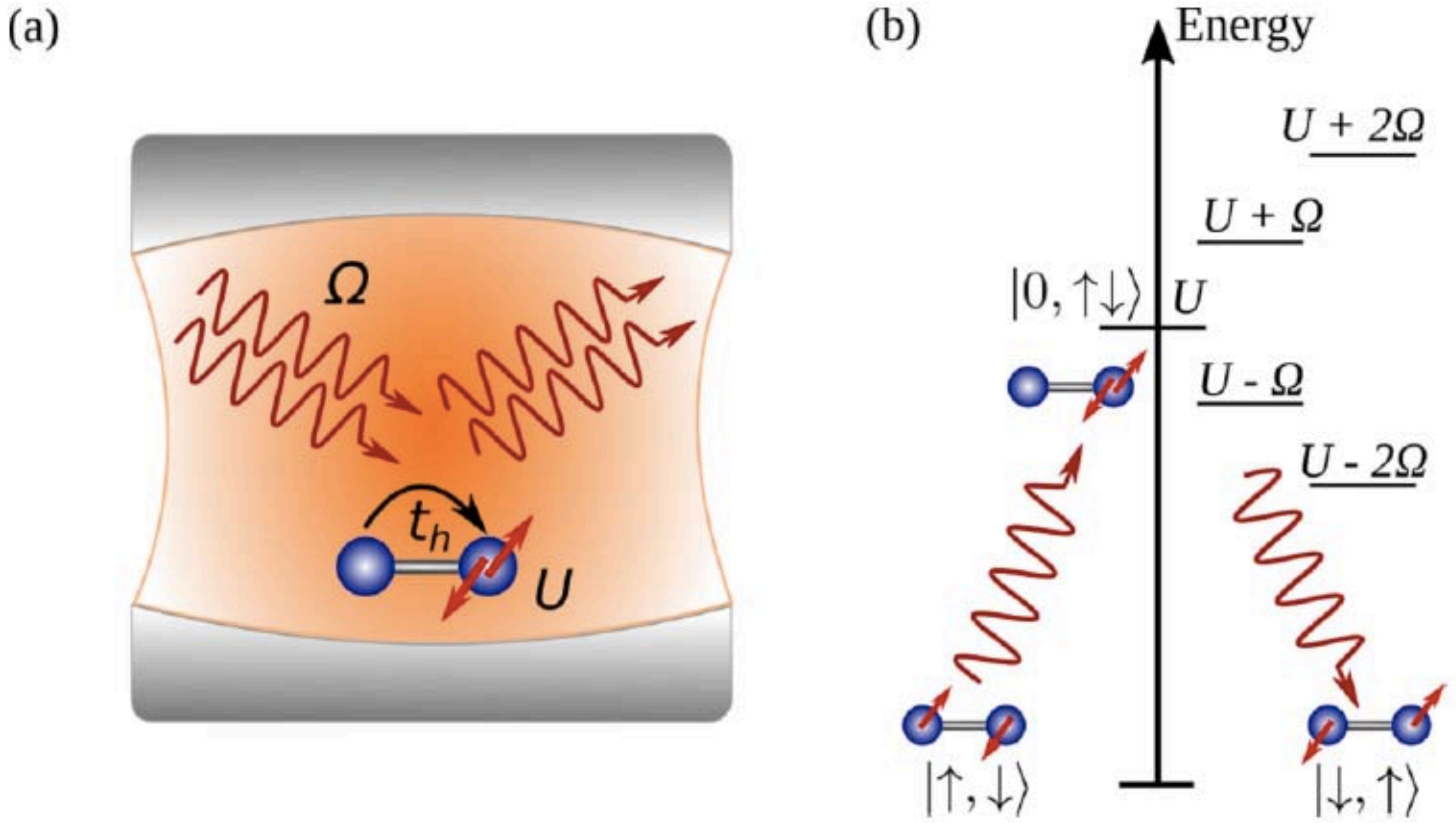
Photon dressing of intermediate states modifies kinetic exchange

But: need for strong lasers, problems with heating, short-lived effect

Question: can we control spin exchange with cavities?
Answer: yes, if we replace strong fields by strong light-matter coupling

Cavity engineering of spin exchange

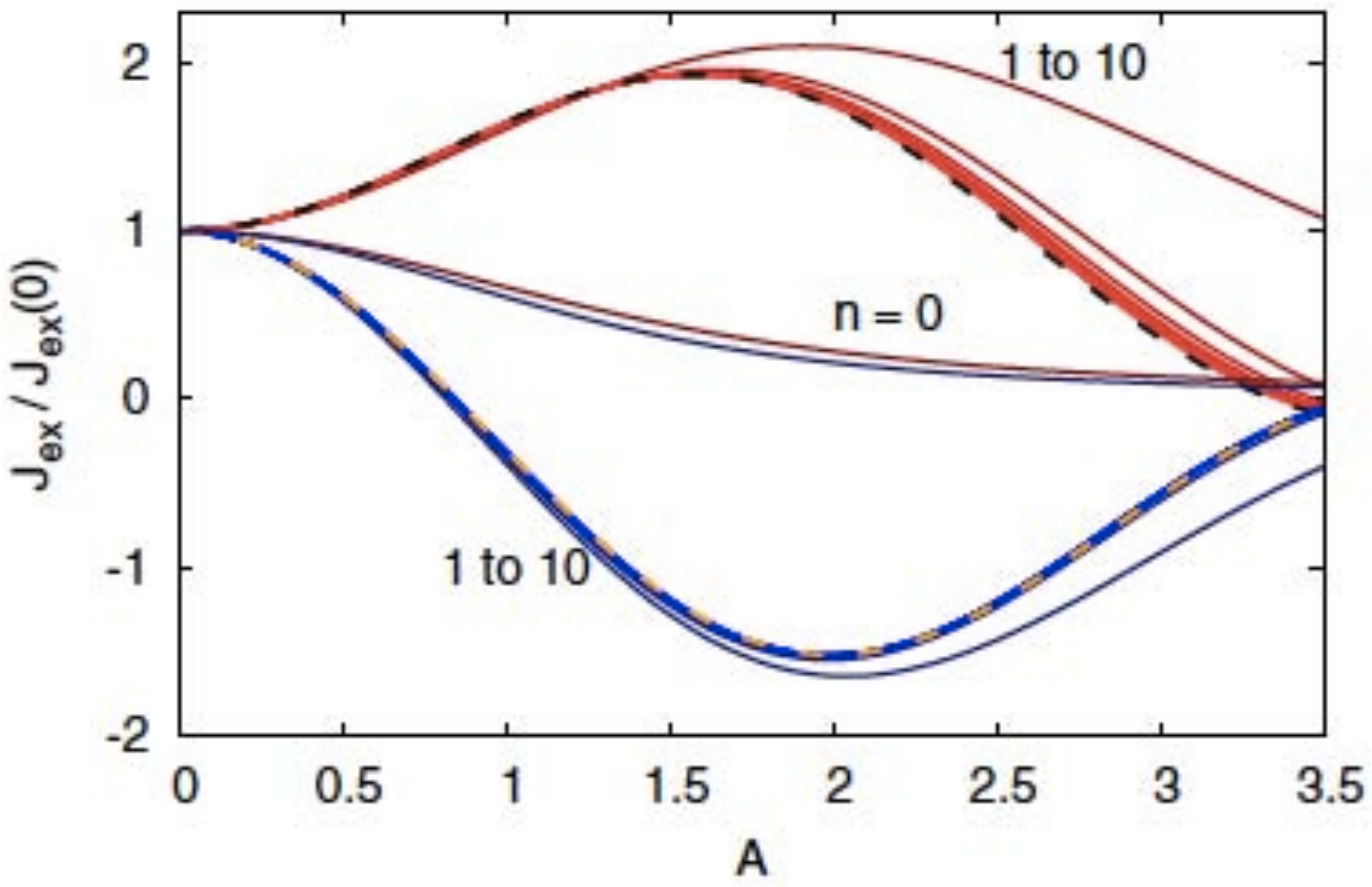
Hubbard model in cavity



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

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

A: effective vector potential
g: light-matter coupling strength



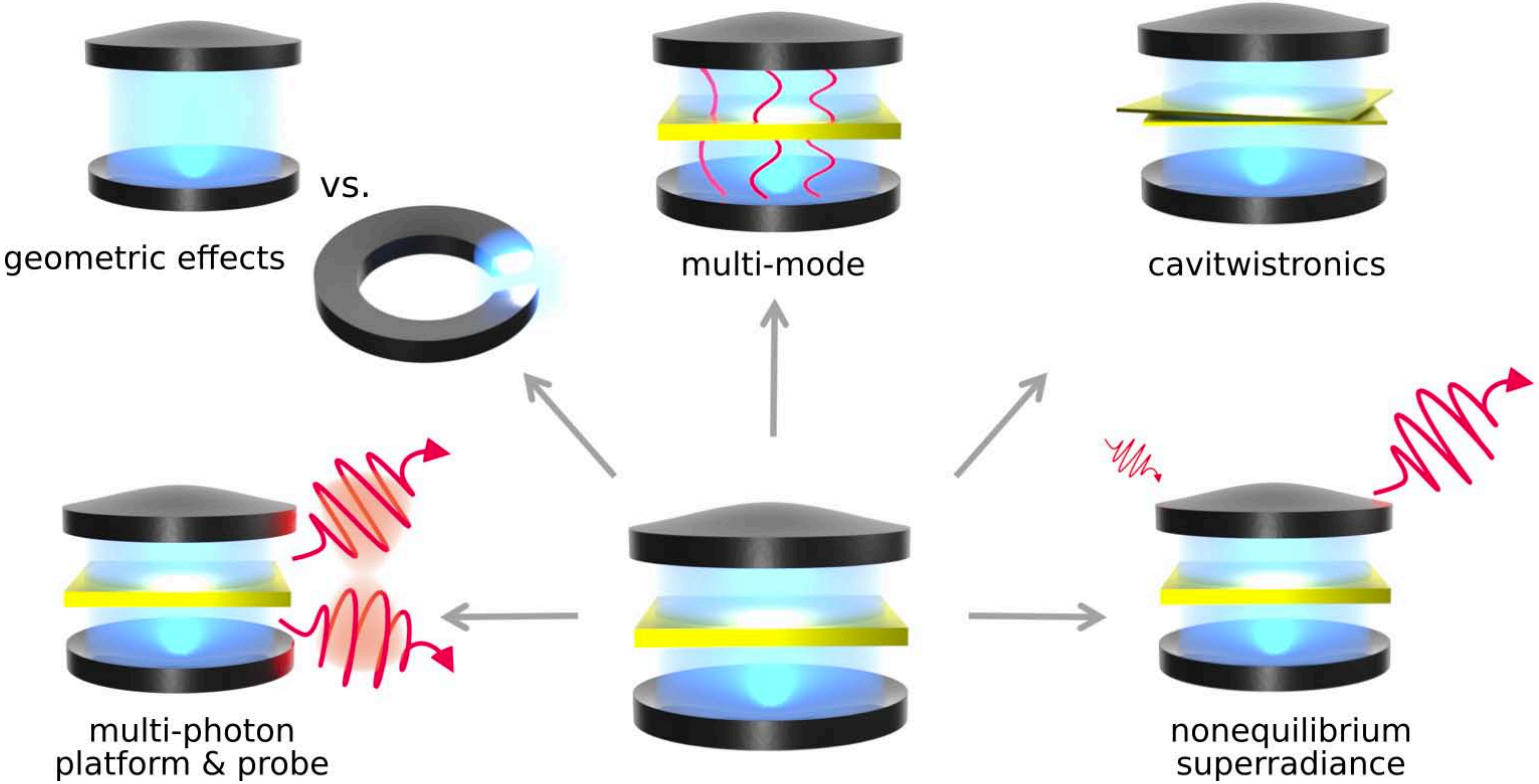
Quantum -> classical Floquet for $n \to \infty, \quad g\sqrt{n}$ fixed.
(large photon number, weak light-matter coupling strength g)

Quantum to classical crossover of Floquet engineering in correlated quantum systems,
M.A. Sentef, J. Li, F. Künzel, M. Eckstein,
PRResearch 2, 033033 (2020)


Jiajun Li Martin Eckstein

Cavity quantum materials

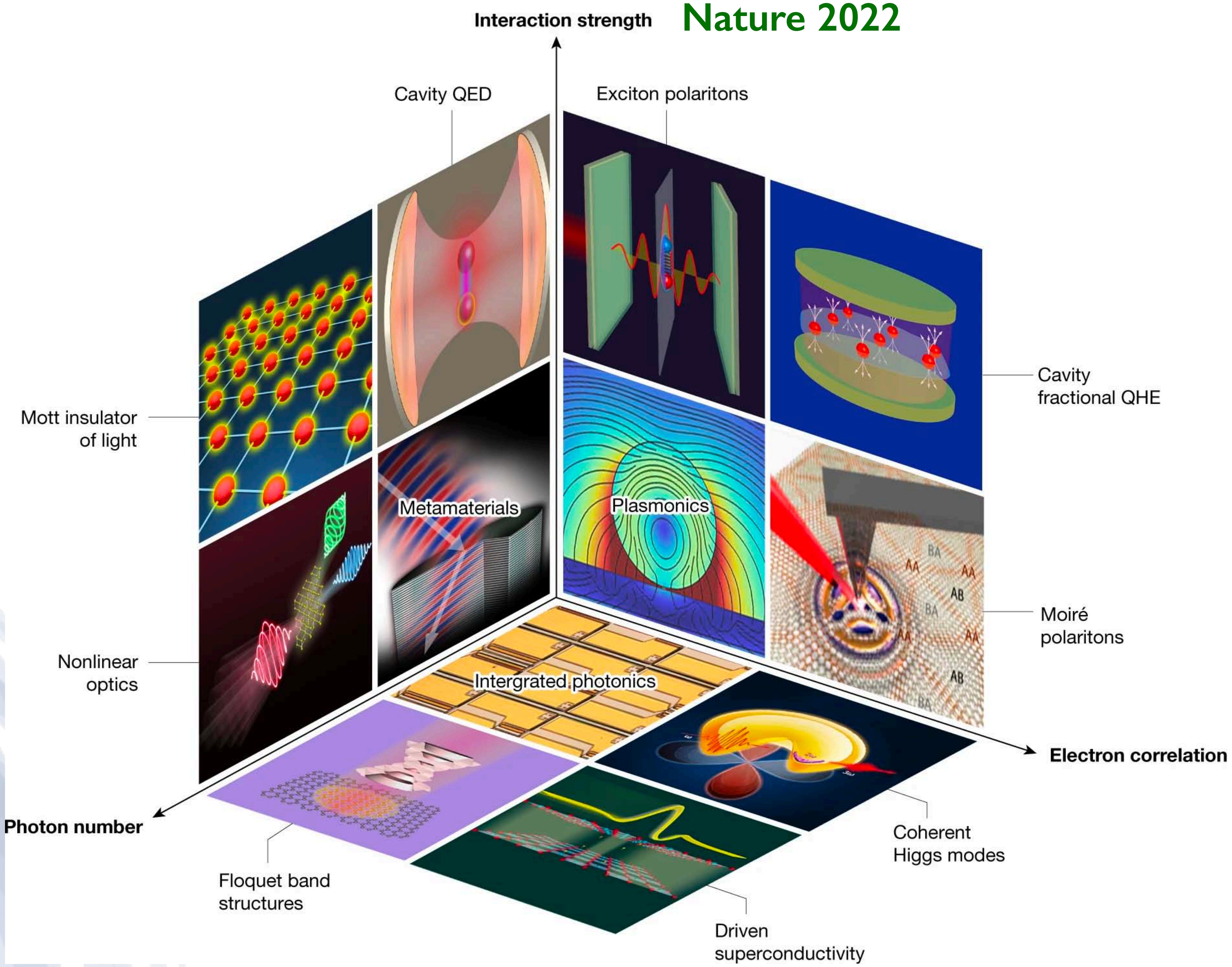
Cavity Quantum Materials, F. Schlawin, D. M. Kennes, MAS,
Applied Physics Reviews 9, 011312 2022



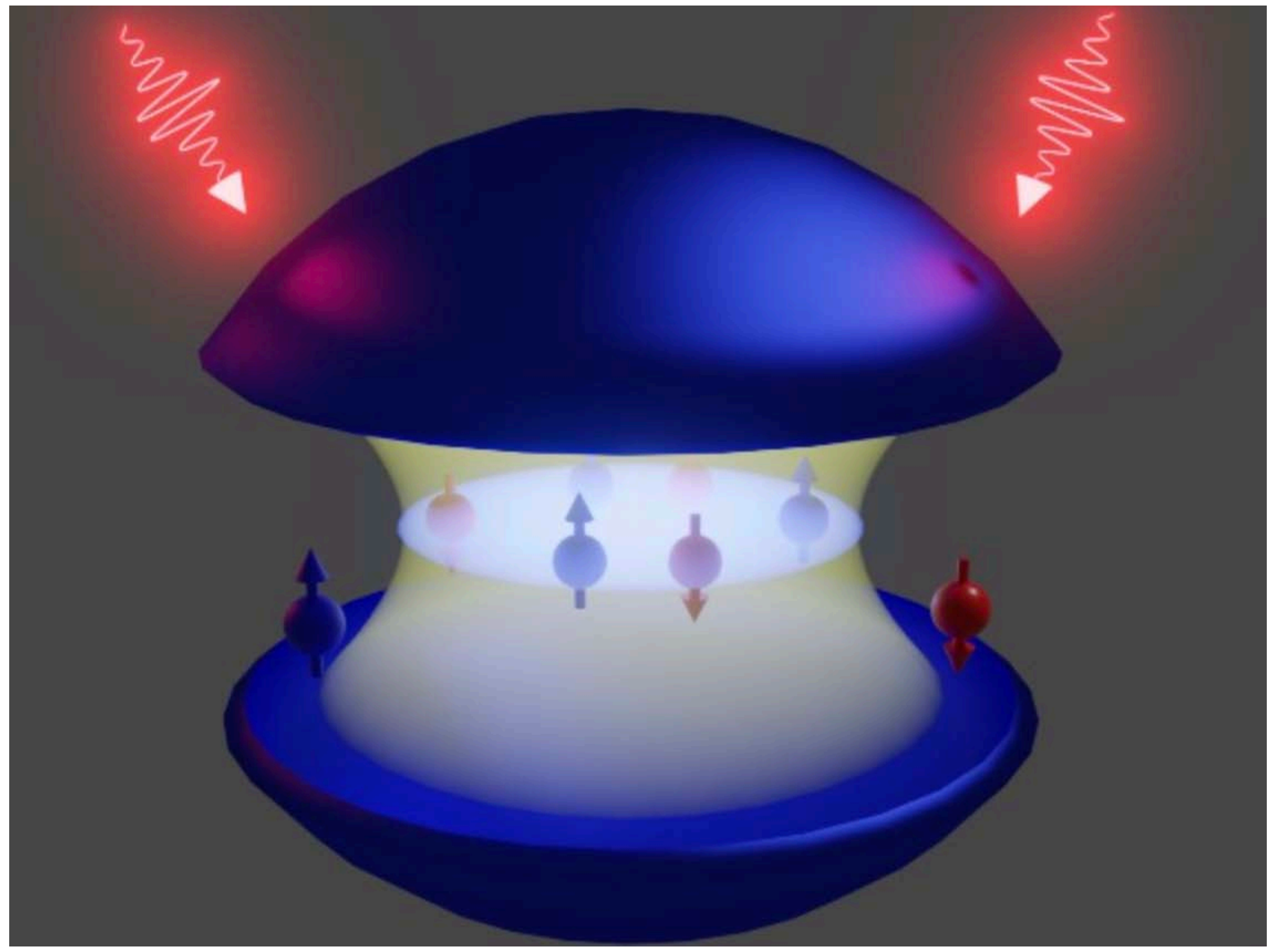
Cavity Quantum Materials, F. Schlawin, D. M. Kennes, MAS, Applied Physics Reviews 9, 011312 2022

Cavity quantum materials

Strongly correlated electron-photon systems, J. Bloch, A. Cavalleri, V. Galitskii, M. Hafezi, A. Rubio, Nature 2022



https://www.kitp.ucsb.edu/activities/qoelectrons25



Quantum Optics of Correlated Electron Systems

Coordinators: Martin Claassen, Mohammad Hafezi, Michael Sentef, and Susanne Yelin

Scientific Advisors: I. Cirac, Eugene Demler, Tony Heinz, and Atac Imamoglu

KITP Workshop Jan/Feb 2025

Summary

Thank you for your attention!

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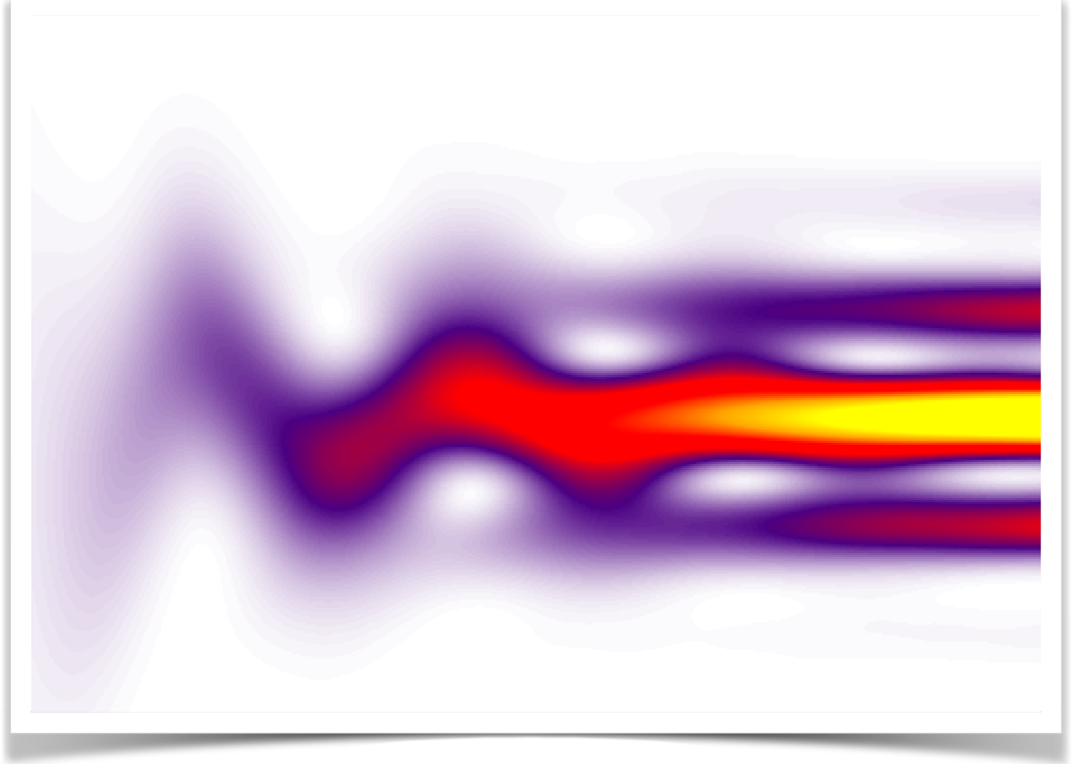
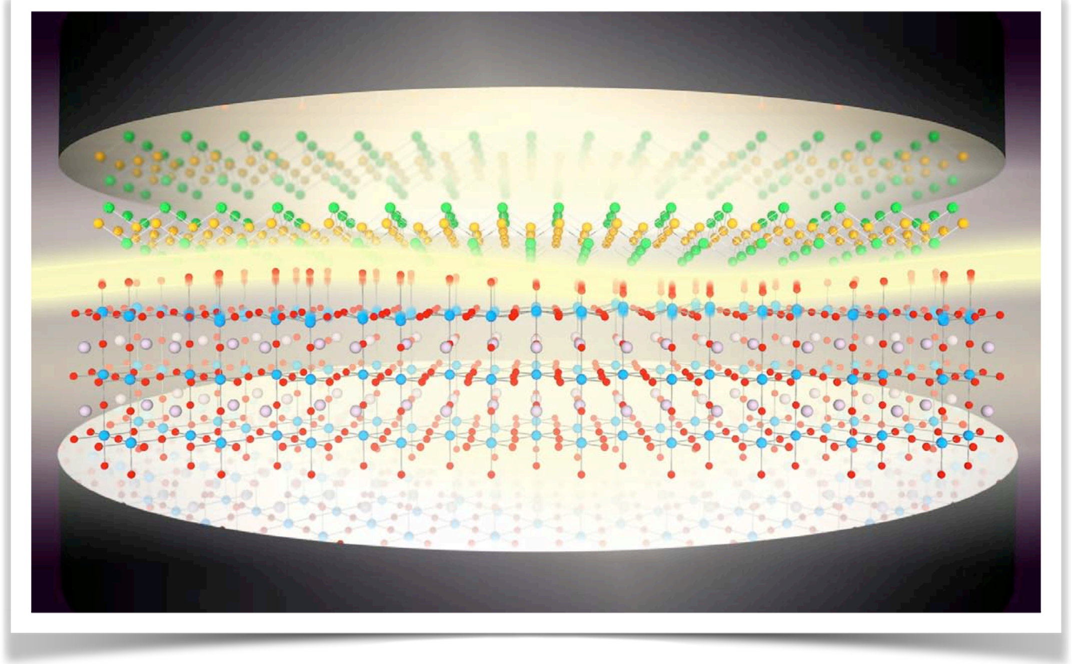


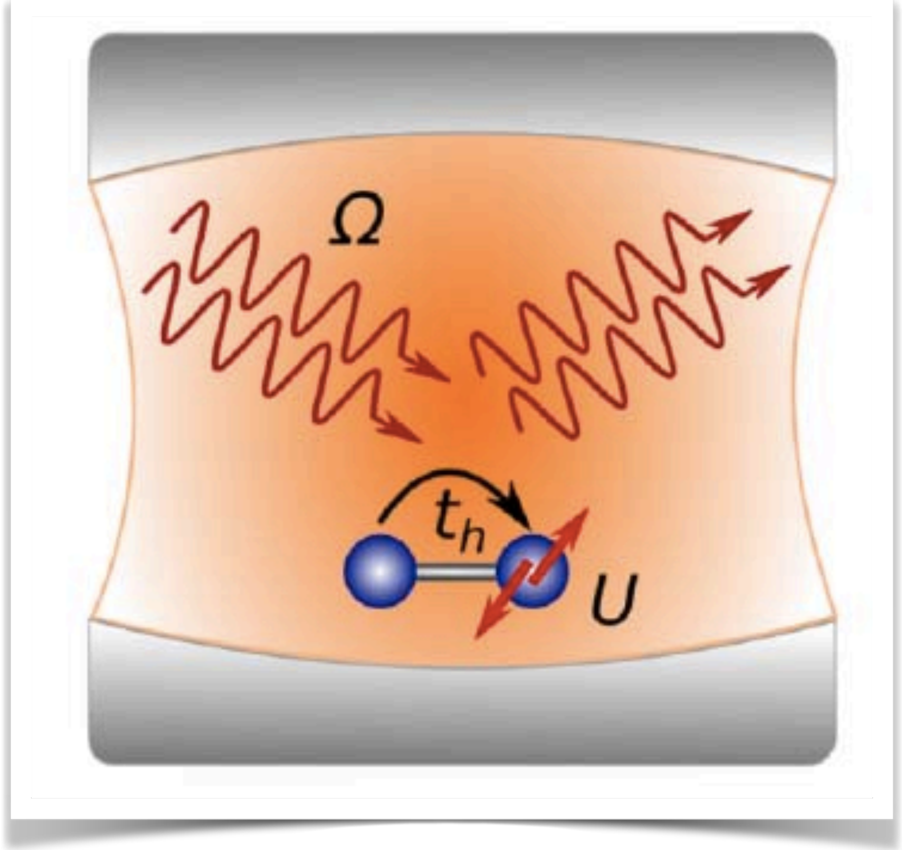
birth of Floquet-Bloch states on subcycle time scales

*Ito, Schüler, et al., Nature **616**, 696–701 (2023)*



cavity quantum materials

*Schlawin, Kennes, Sentef, Applied Physics Reviews **9**, 011312 (2022)*



cavity engineering of topology and correlations

*Wang, Ronca, Sentef, Phys. Rev. B **99**, 235156 (2019)*
*Sentef, Li, Künzel, Eckstein, Phys. Rev. Research **2**, 033033 (2020)*