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Can we manipulate quantum materials via strong light-matter coupling in cavities?

Martin Eckstein

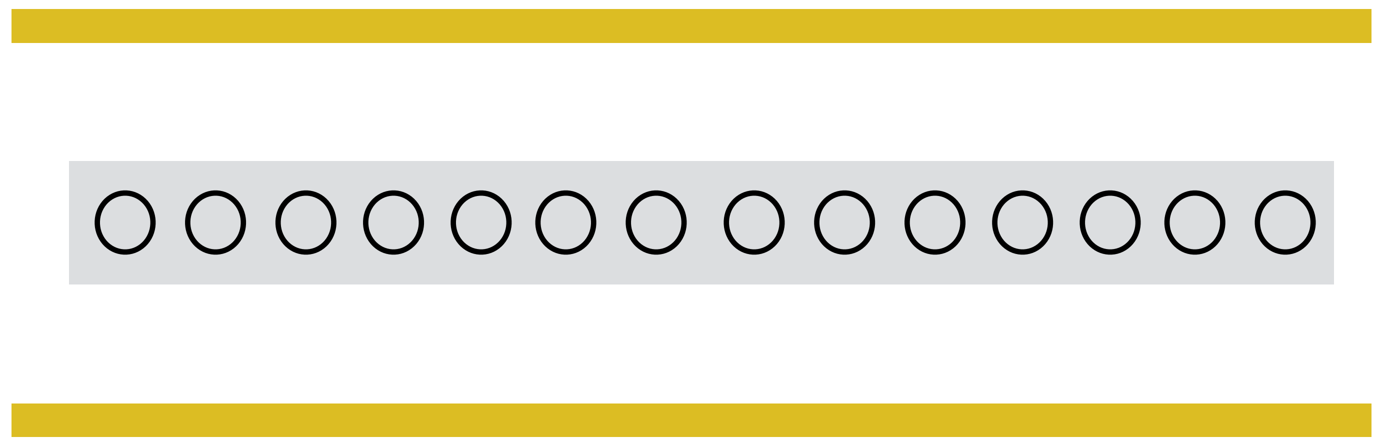
SPICE workshop “Quantum Spinoptics”, June 14, 2023

Deutsche Forschungsgemeinschaft

erc

TRR 306
QuCoLiMa
Quantum Cooperativity of Light and Matter

“Cavity quantum materials”



Engineer coupling of an extended solid to the *vacuum fluctuations* of the electromagnetic field to manipulate ground state and thermodynamic properties?

… see also talk by J. Kono

Review/Perspective: Schlawin, Kennes, Sentef, App. Phys. Rev. **9**, 011312 (2022)
Bloch Cavalleri, Galitski, Hafezi, Rubio, Nature **606,** 41 (2022)

“Cavity quantum materials”

Schlawin, Kennes, Sentef, App. Phys. Rev. **9**, 011312 (2022)
Bloch Cavalleri, Galitski, Hafezi, Rubio, Nature **606,** 41 (2022)

Possible mechanisms:

1) Light-mediated long-range interactions

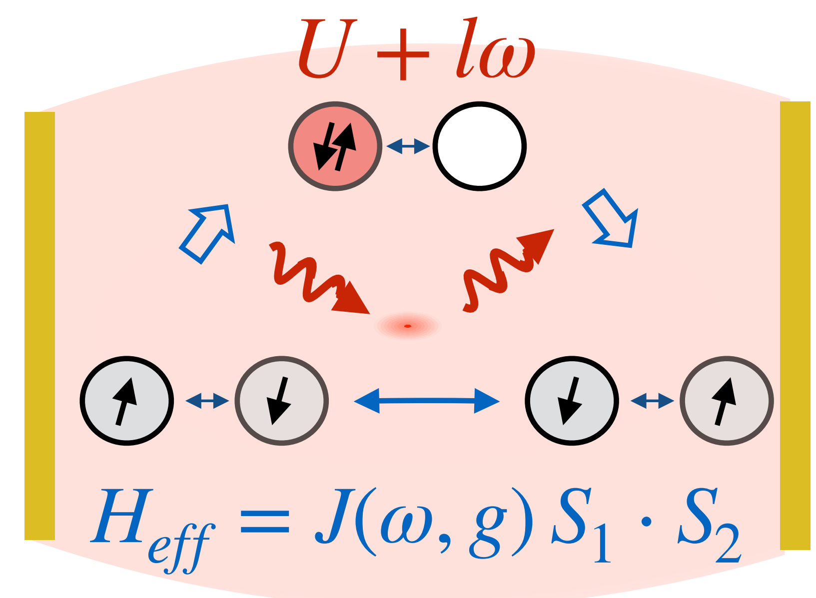
Polariton mediated superconductivity
Photon-mediated superconductivity …

2) Single-particle effects:

- Light-dressed quasiparticles (electron-polariton band structures)
- Light-dressed local interactions

e.g.: photon-dressed superexchange
Kiffner et al. (2019)

close link to Floquet Hamiltonian
Sentef, Künzel, Li, Eckstein, PRR 2020



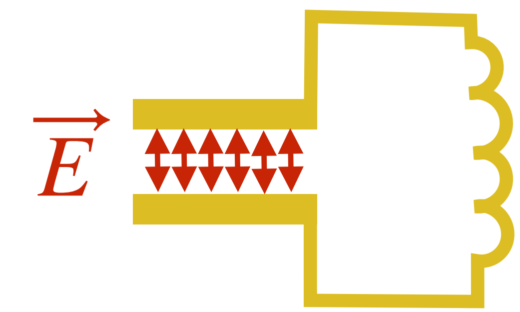
Variations:

Superconducting pairing & CDW interactions — Li, ME, PRL **125**, 217402 (2020)
Frustrated spin models — Boström et al., arXiv:2211.07247
Magnetic impurities in SC — Müller, ME & Viola-Kusminskiy, PRL 2023

Strong light-matter coupling in the thermodynamic limit?

General issue:

single mode resonator ⇨ single particle coupling $g \propto V^{-1/2}$



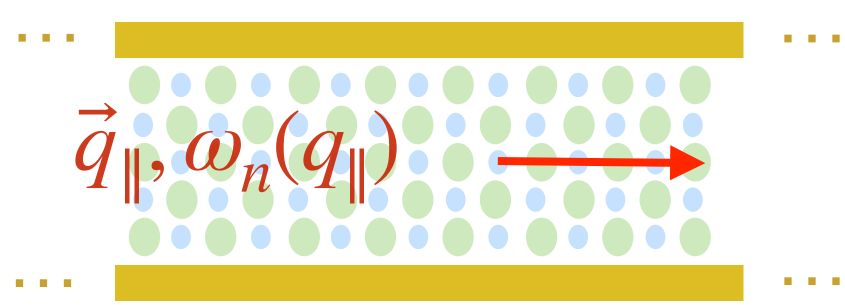
⇨ no single-particle strong coupling effects in thermodynamic limit

⇨ Only “collective effects” depending on N/V survive (Rabi splitting)

Path to thermodynamic limit?

1) Can collective coupling to a single mode affect *equilibrium* thermodynamics?
Condensation of hybrid light-matter modes (“superradiance”)?

2) Extended cavity

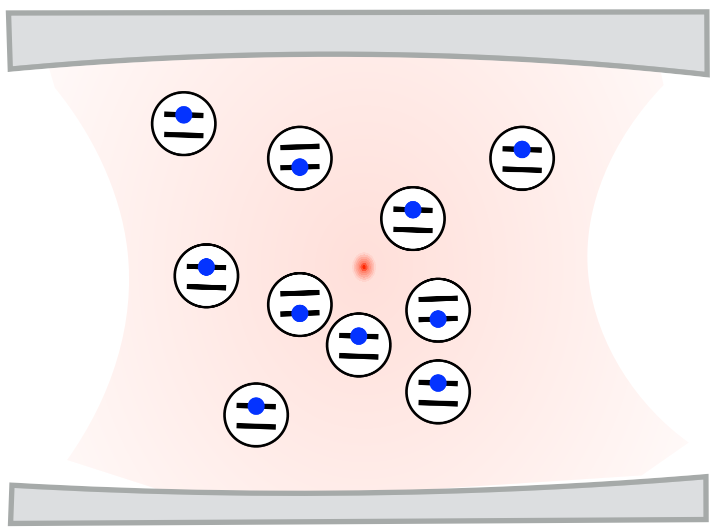


Effect on solid must be understood due to coupling to **mode continuum**

E.g.: Resummation of quantum Floquet Hamiltonian: Li et al., PRB, **105**, 165121 (2022)

Single mode collective coupling

Minimal model: Dicke model



$$\vec{E} \sim \hat{X}$$

$$H = \Delta \sum_{j=1}^{N} \sigma_{j,z} + \frac{\Omega}{2}\left(P^2 + X^2\right) + \frac{g}{\sqrt{V}}\sqrt{\Omega} \sum_j \hat{\sigma}_{j,x} X$$

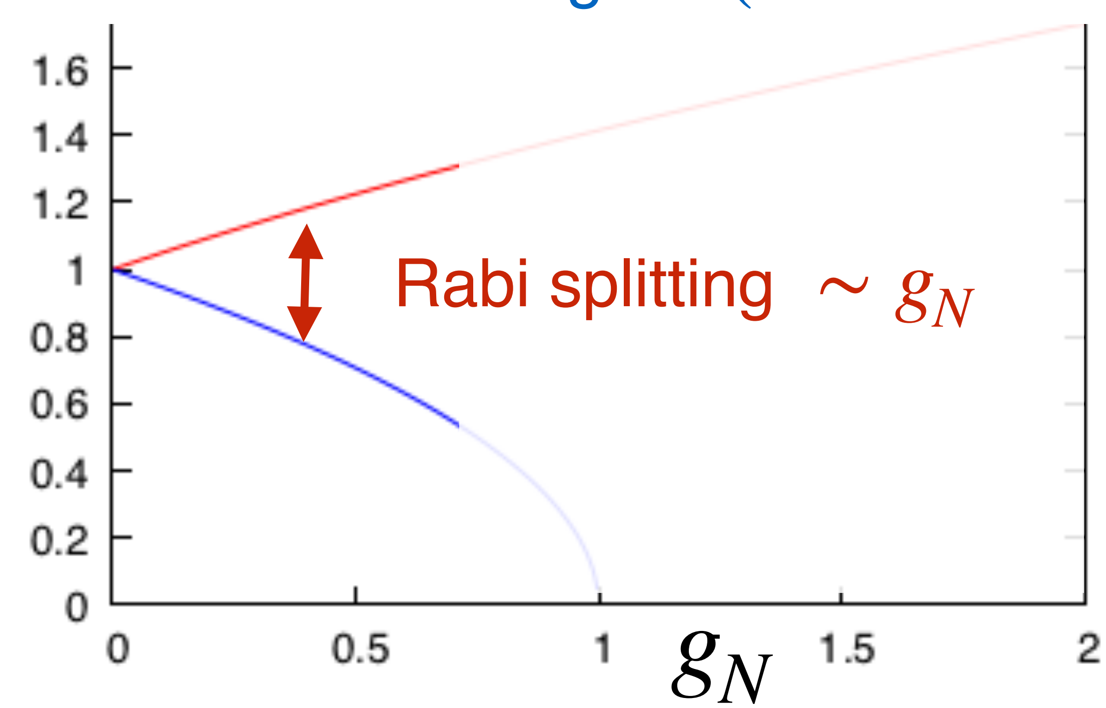
cavity mode

- $N, V \to \infty \quad N/V$ fixed ⇨ theory for collective dipole $\hat{Q} = \frac{1}{\sqrt{N}} \sum_j \hat{\sigma}_{j,x}$

$$H_{coll} = \frac{\Delta}{2}\left(P_Q^2 + Q^2\right) + \frac{\Omega}{2}\left(P^2 + X^2\right) + g_N \sqrt{\Omega}\, XQ$$

collective coupling $g_N \sim \sqrt{N/V}$

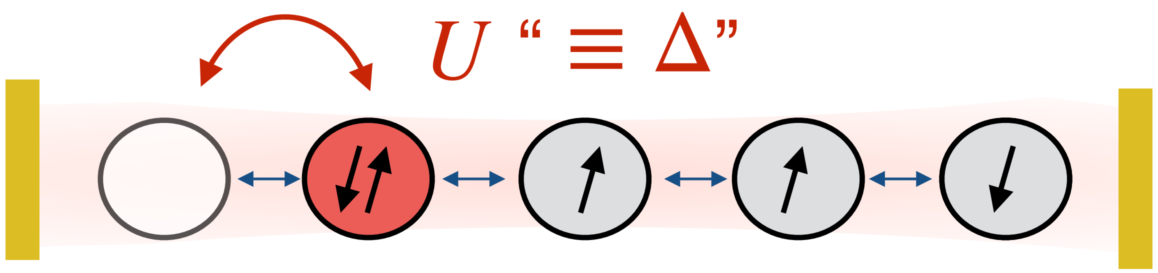
Normal mode energies $(\Delta = \Omega = 1)$



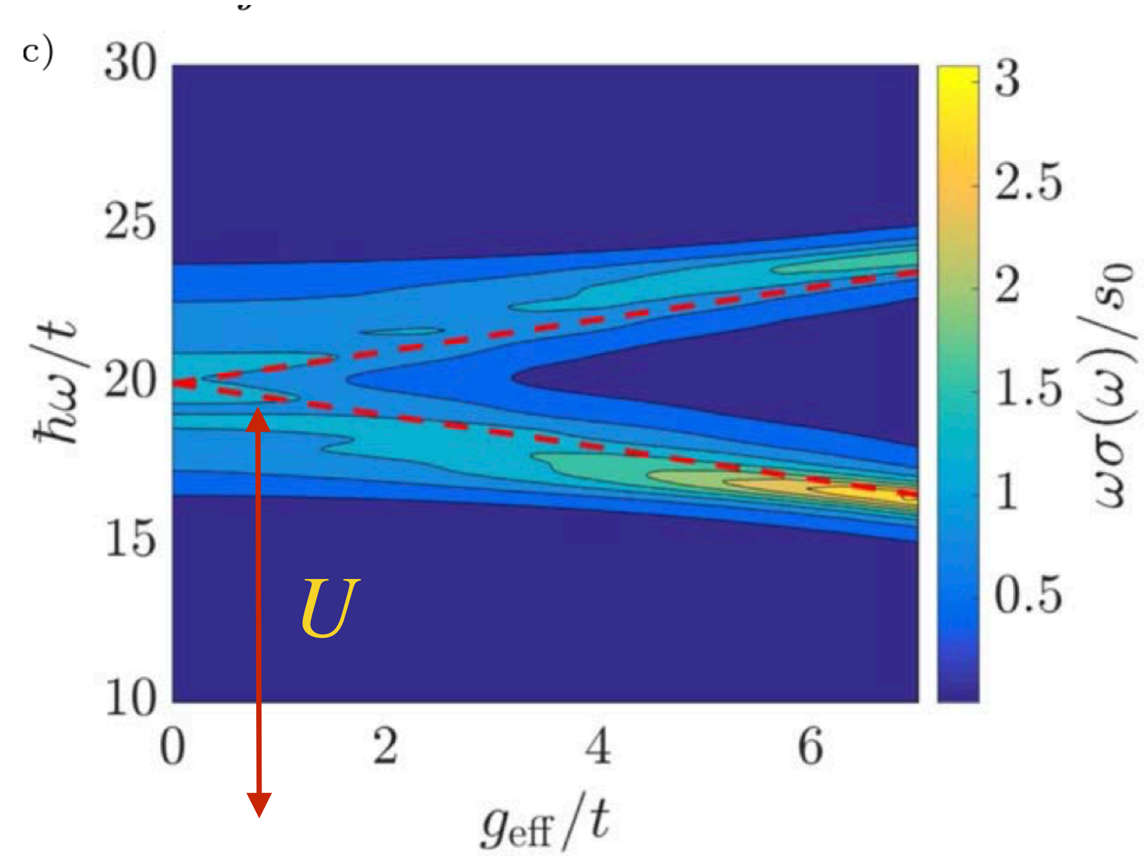
Single mode collective coupling

$g_N \sim \sqrt{N/V}$ ⇨ ultra-strong collective coupling in solid accessible … talk by J. Kono!

E.g.: Theoretical solution of extended Hubbard model (ET-F_2TCNQ) in cavity:



Martin Kiffner et al. New J. Phys. **21** 073066 (2019)



But:

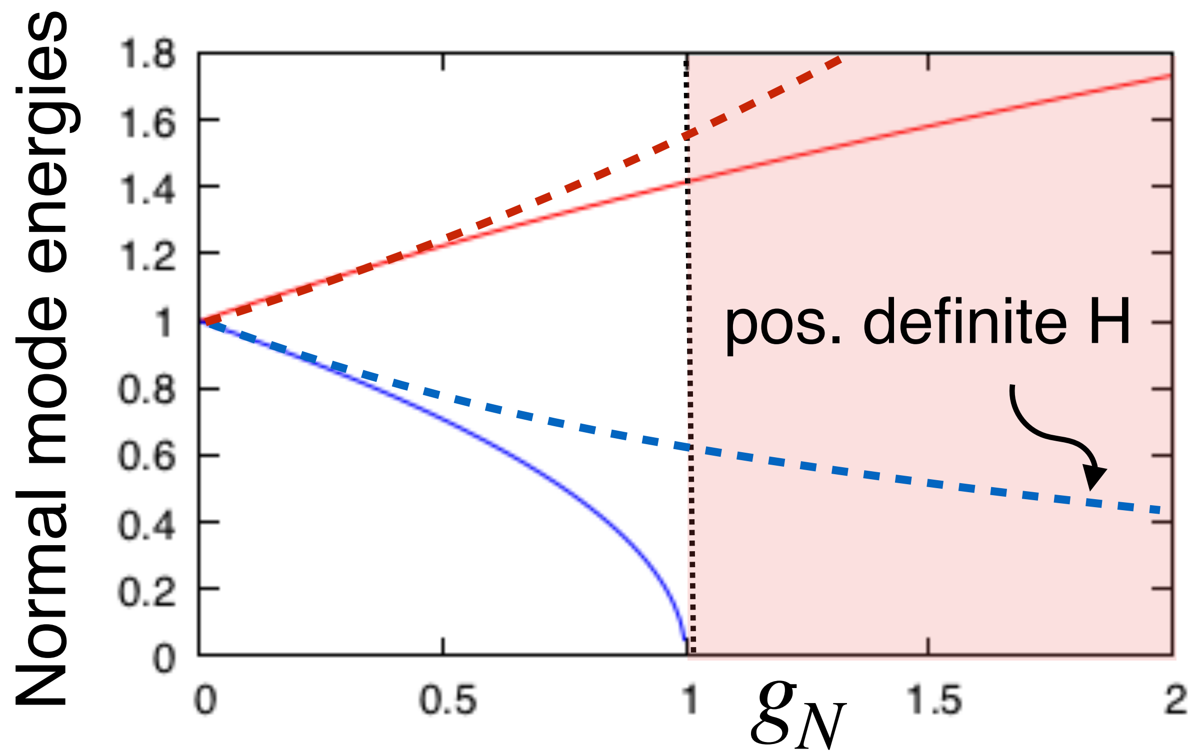
- classical explanation using free-space conductivity $\sigma(\omega)$ & macroscopic Maxwell

(with sub-leading corrections in terms of nonlinear free space response $\chi^{(4)}, \chi^{(6)}, \ldots$)
Lenk, Li, Werner, Eckstein, arXiv:2205.05559

⇨ Collective Rabi splitting ≠ ground state quantum light-matter hybrid!

… unless mode would condense

(Equilibrium) Dicke superradiance



⇨ Instability for $g_N > 1$

Hepp and Lieb, (1973)

$\langle a \rangle \neq 0$: "superradiant transition"

$\langle \sigma_x \rangle \neq 0$: ferroelectric transition

$$H = \frac{\Delta}{2}\left(P_Q^2 + Q^2\right) + \frac{\Omega}{2}\left(P^2 + X^2\right) + g_N\sqrt{\Omega}\,XQ + \frac{g_N^2}{2}Q^2$$

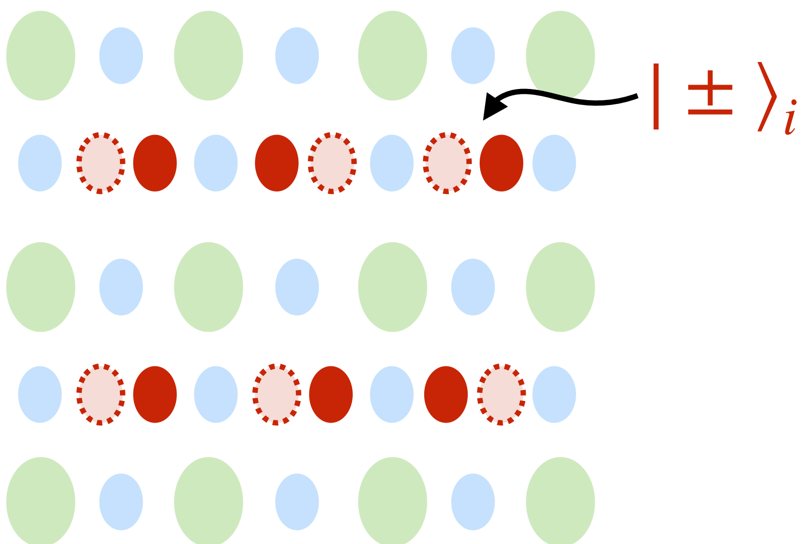
positive definite $\frac{\Omega}{2}\left(X + g_N Q/\sqrt{\Omega}\right)^2$

from positive minimal coupling "$(p-A)^2$"

⇨ absence of phase transition "no go theorem" Rzazewski, et al. PRL **35**, 432 (1975)

- Re Here: Minimal model which shows how coupling to **mode continuum** can assist the condensation of the ferroelectric mode (superradiance)

Transverse field Ising model for "Quantum para-electric" (QPE)

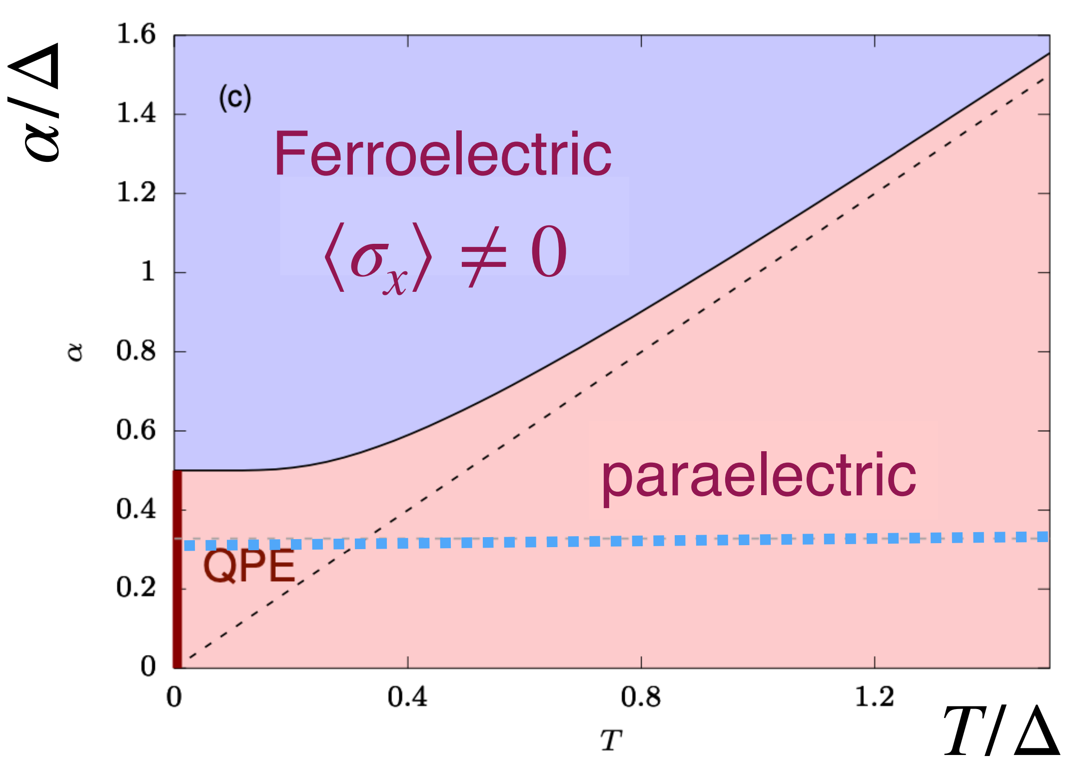


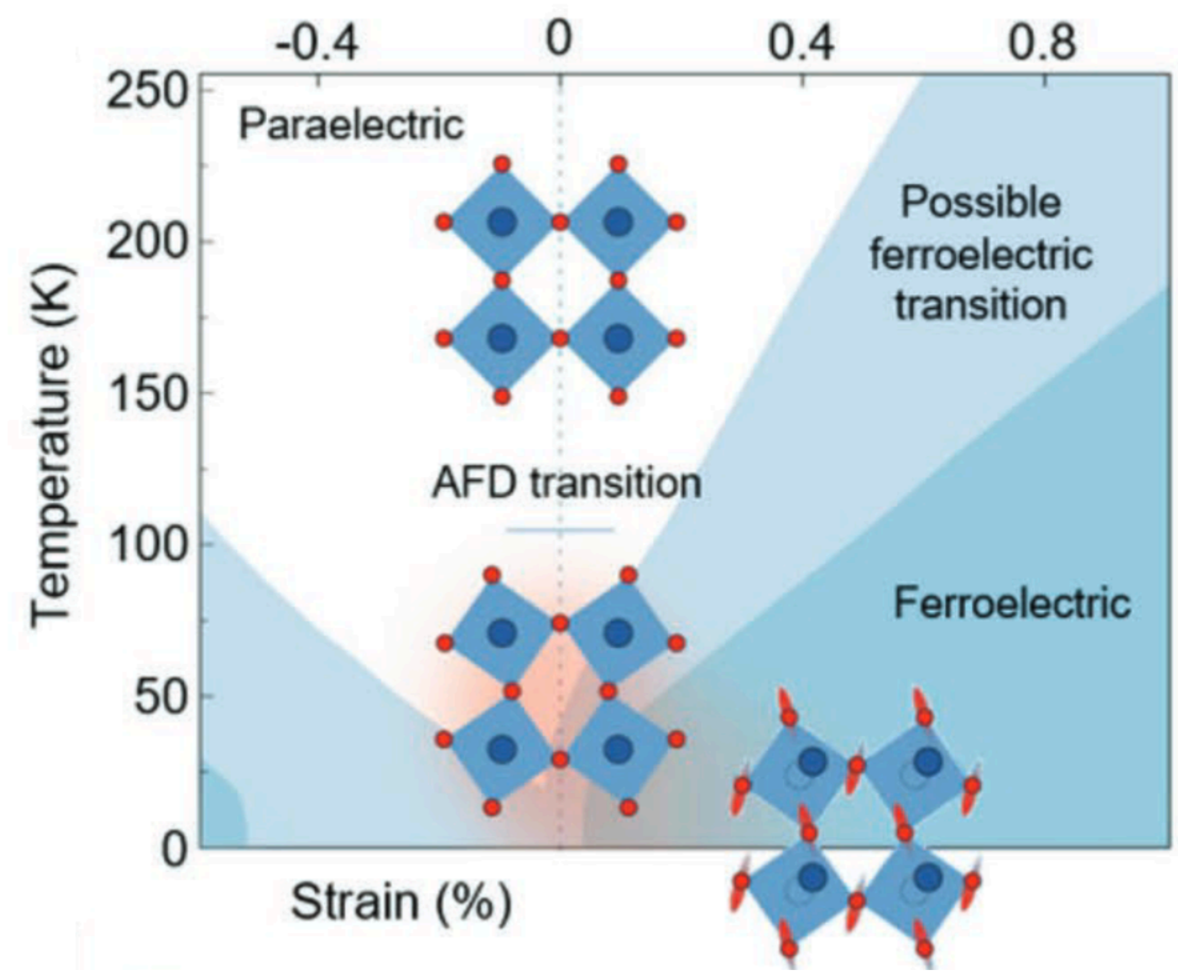
$$H_{solid} = \frac{\Delta}{2}\sum_{j=1}^{N}\sigma_{j,z} \quad - \quad \frac{\alpha}{4}\sum_{\langle j,j'\rangle}\sigma_{j,x}\sigma_{j',x}$$

Tunneling
$|+\rangle \leftrightarrow |-\rangle$

N interacting dipoles

mean-field phase diagram:





From Nova et al. Science (2019)

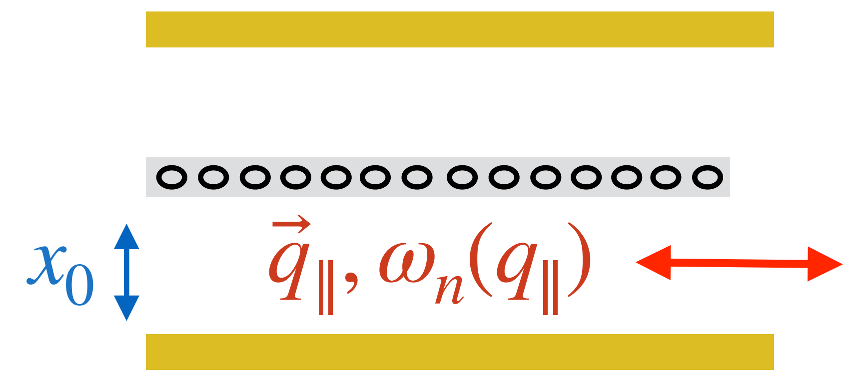
quantum fluctuations

ferroelectricity

Transverse field Ising model for “Quantum para-electric”

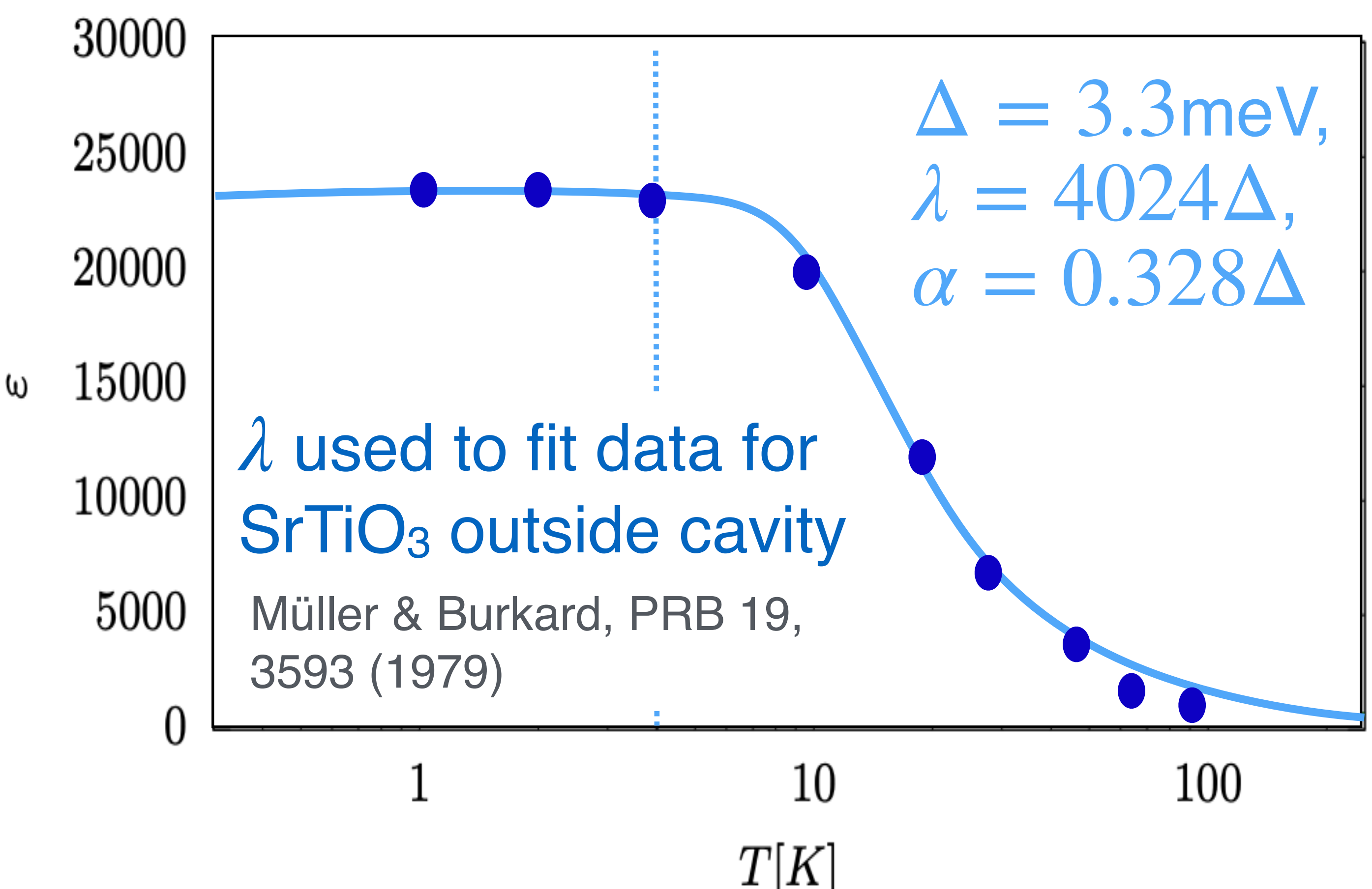
Full Hamiltonian:

$$H = H_{solid} + \sum_{q_{||}} \omega_{q_{||}} a^{\dagger}_{q_{||}} a_{q_{||}} + H_{EP} + H_{PP}$$



Linear coupling to field

$$H_{EP} = \sum_j \sigma_{j,x}\, e\vec{d} \cdot \vec{E}(\vec{r}_j) = \sum_{j,q_{||}} \sqrt{\frac{\omega_{q_{||}}}{2N}} \left[g_{q_{||}} a_{q_{||}} e^{iq_{||}r_j} + h.c. \right] \sigma_{j,x}$$



Set by shape of mode-function

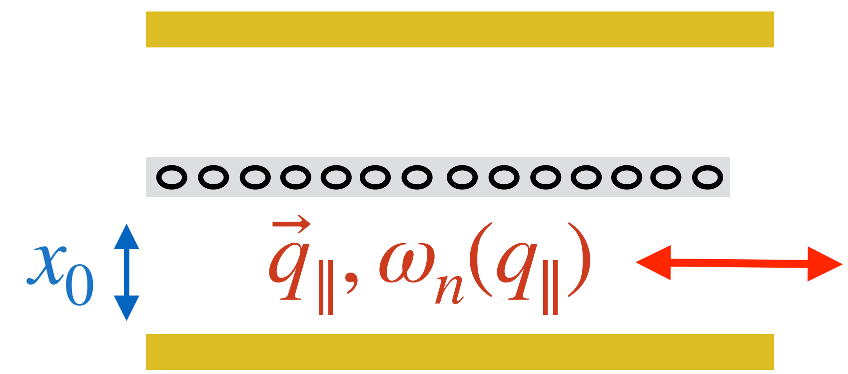
$$|g_{q_{||}}|^2 = \lambda \, | u_{q_{||}}(x_0) \cdot e_d |^2$$

$$\lambda = \frac{e^2 d^2}{\epsilon_0 a^3}$$

Transverse field Ising model for "Quantum para-electric"

Full Hamiltonian:

$$H = H_{solid} + \sum_{q_{||}} \omega_{q_{||}} a^{\dagger}_{q_{||}} a_{q_{||}} + H_{EP} + H_{PP}$$



Linear coupling to field

$$H_{EP} = \sum_{j} \sigma_{j,x}\, e\vec{d} \cdot \vec{E}(\vec{r}_j) \;\; = \sum_{j,q_{||}} \sqrt{\frac{\omega_{q_{||}}}{2N}} \left[g_{q_{||}} a_{q_{||}} e^{iq_{||}r_j} + h.c. \right] \sigma_{j,x}$$

$$H_{PP} = \sum_{j,j'} \sum_{q_{||}} \frac{|g_{q_{||}}|^2}{2N} e^{iq_{||}(r_j - r_{j'})} \sigma_{j,x} \sigma_{j',x}$$

… so that $H_{field} + H_{EP} + H_{PP}$ is posititive definite

Induced interactions

Integrate out photon $\rightarrow$ **Cavity-induced interaction** $V_{ij}(\omega)$ $\propto \omega^2$

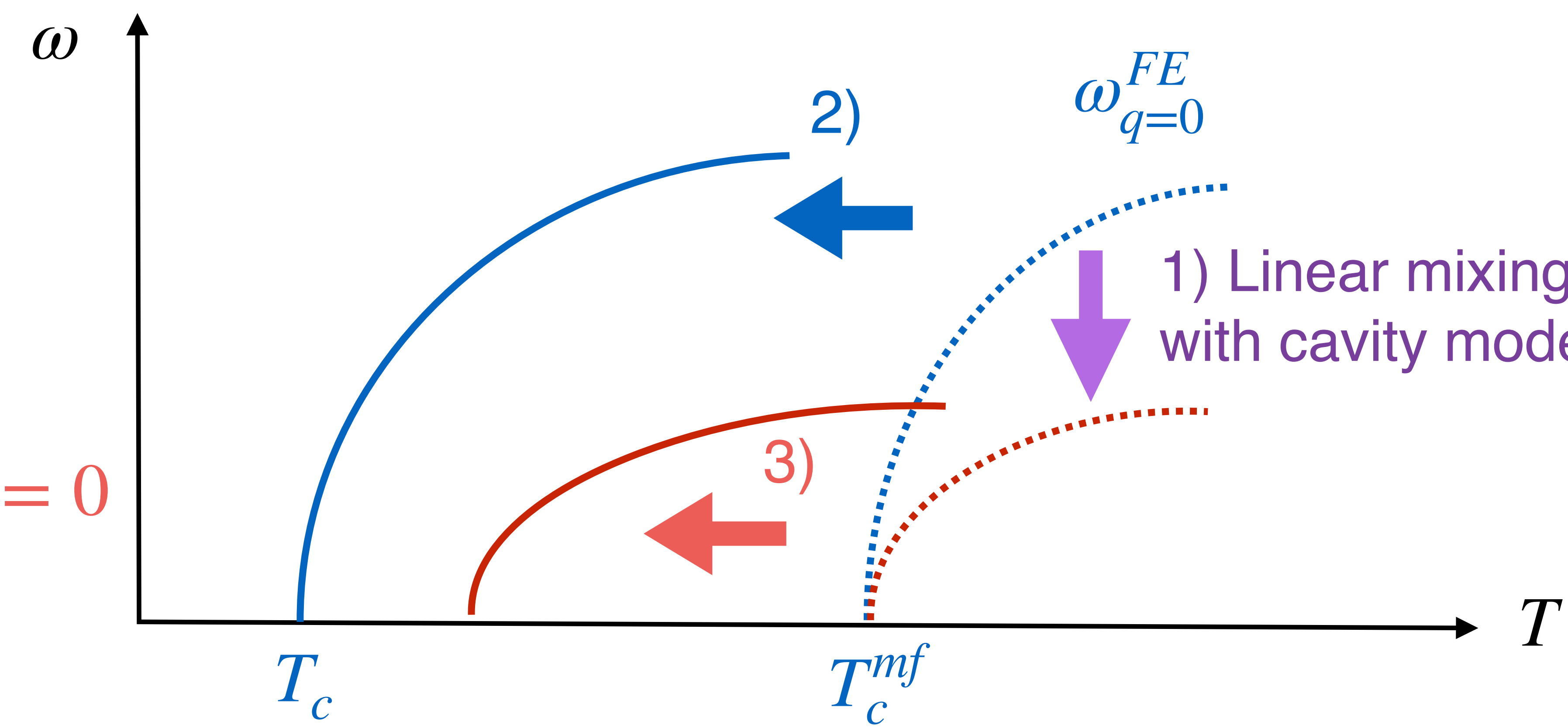
$$V_q(\omega) = \underbrace{|g_q|^2}_{\sigma_j^x \sigma_j^x \text{ term in } H_{PP}} + \underbrace{|g_q|^2 \frac{\omega_q^2}{\omega^2 - \omega_q^2}}_{\text{eliminating photon}} = |g_q|^2 \frac{\omega^2}{\omega^2 - \omega_q^2}$$

$$V_{ind}(\omega = 0) = 0$$ ⇨ No effect of cavity in static mean-field theory!

Phenomenological theory beyond mean field: Ashida et al. PRX (2020)

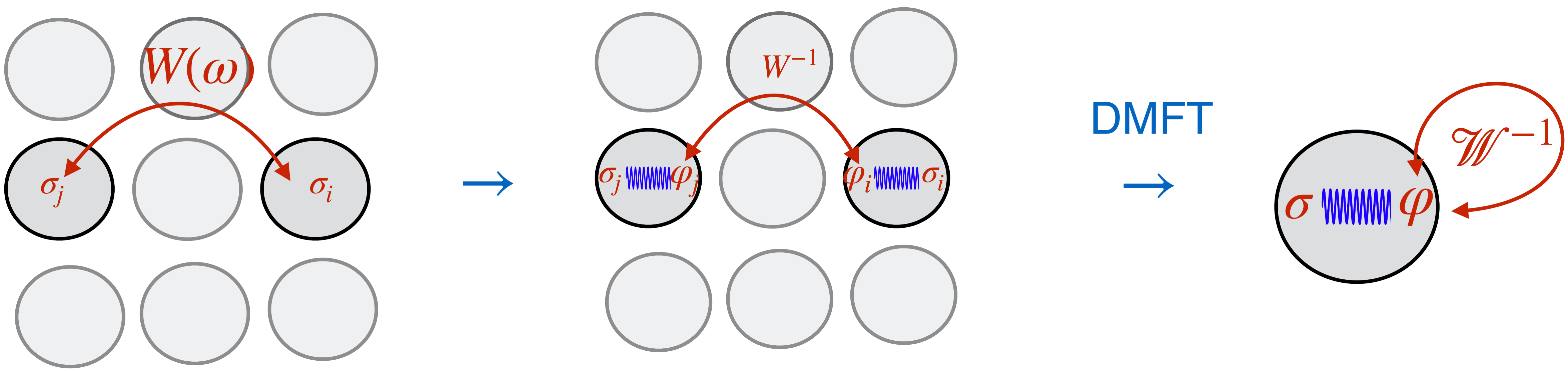
2) Renormalization of T_c by anharmonic interaction $q = 0 \leftrightarrow q \neq 0$:

3) Anharmonic interaction $q = 0$ with **hybrid** modes $q \neq 0$



Microscopic treatment beyond mean field

⇨ Solution by dynamical mean field theory: Georges et al., RMP 1996

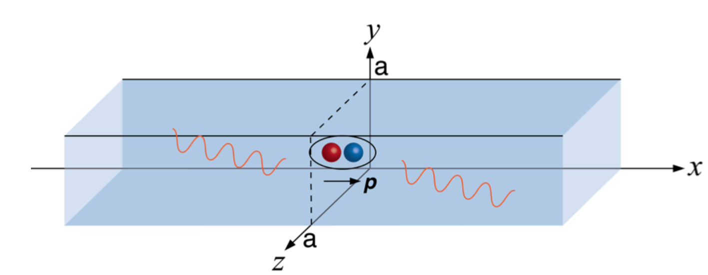


W: full interaction
(Ising & induced)

Harmonic theory for real field φ with **local** interaction

single-site in self-consistent environment

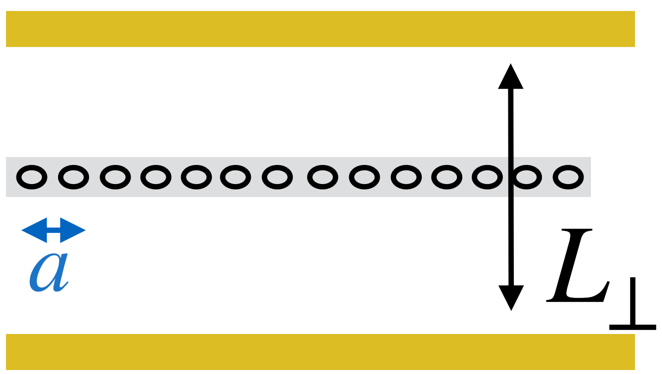
Atom (two level system) with strong coupling to “effective continuum” (∼ waveguide QED)



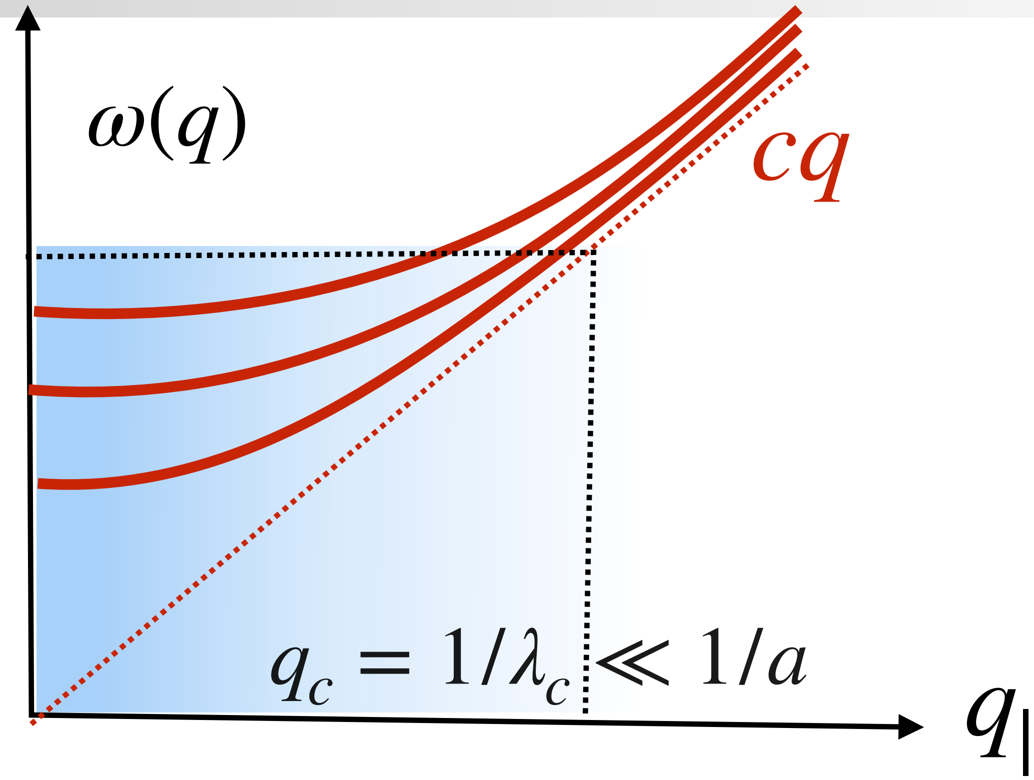
⇨ Diag-QMC based on expansion in retarded spin interaction Kim et al., PRL **130**, 036901 (2023)

See also: DMFT for plain φ^4-theory: Akerlund et al., PRD **88**, 125006 (2013)

How strong is induced interaction?



$$\omega_c = \frac{\pi}{L_\perp c}$$



$1/N_{cells} \sim a^3/V$

Induced interaction $V_{ij} = \sum_q e^{iq(R_i - R_j)} |g_q|^2 \frac{\omega^2}{\omega^2 - \omega_q^2}$

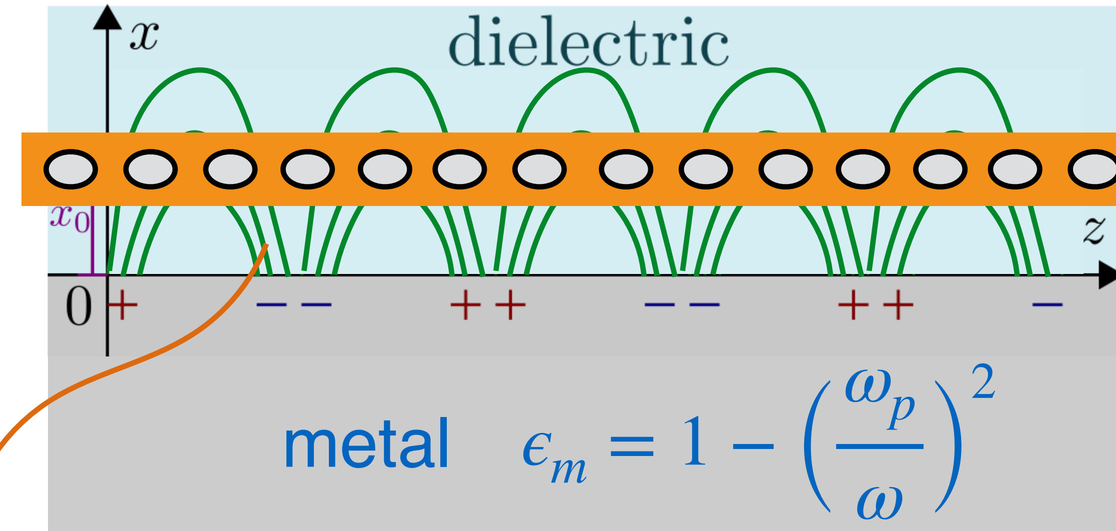
Cutoff: only modes which are affected by the cavity should be included $\sim V/\lambda_c^3 \ll N_{cells}$

Main design challenge for “cavity quantum materials”: overcome phase space restriction!

Electromagnetic field confined at surface:

Surface Plasmon Polariton (SPP) mode:

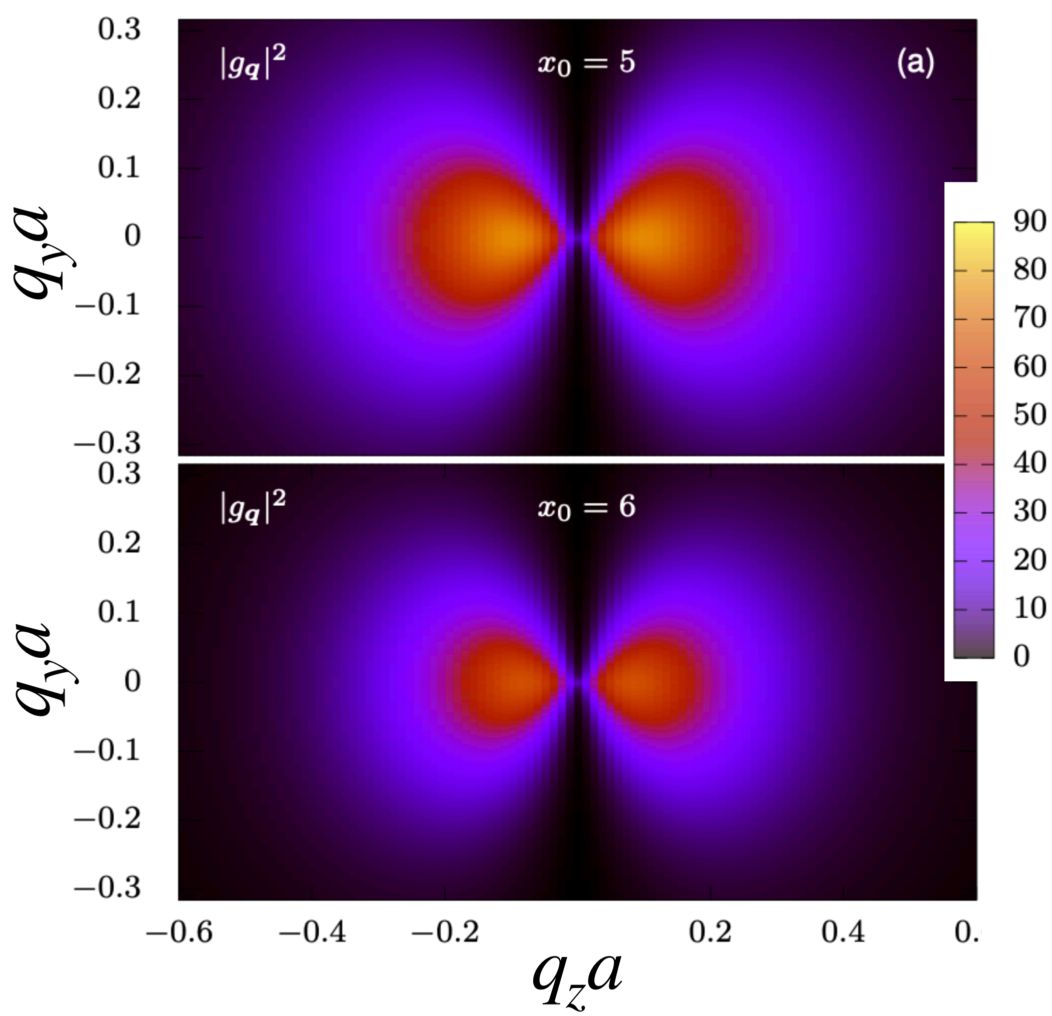
Economou, Phys. Rev. 182, 539 (1969)
Ashida et al. PRX 2020

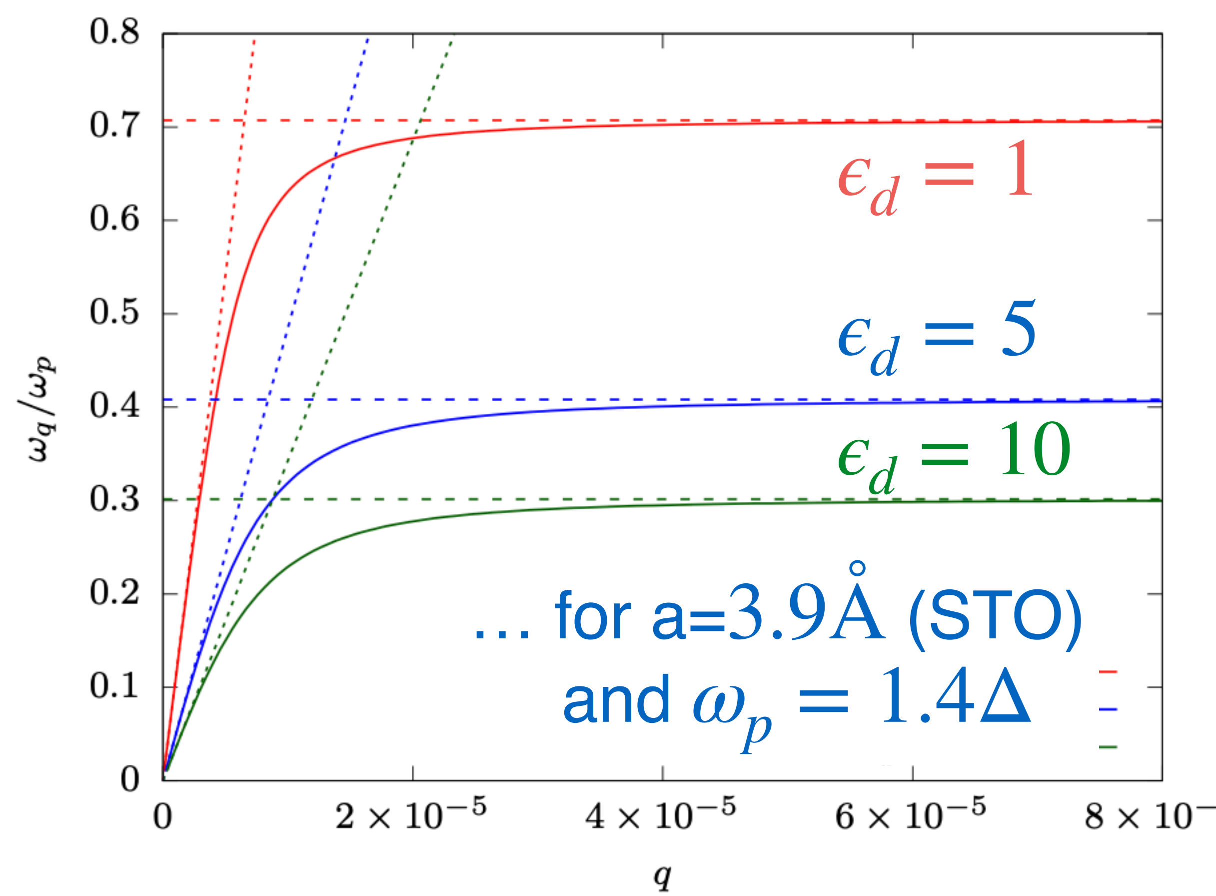


- Flat dispersion ($\omega(q) \to$ const.) for large q
- localization at interface: decay $\sim 1/|q_{||}|$

Setting: 2d material, distance x_0 from surface

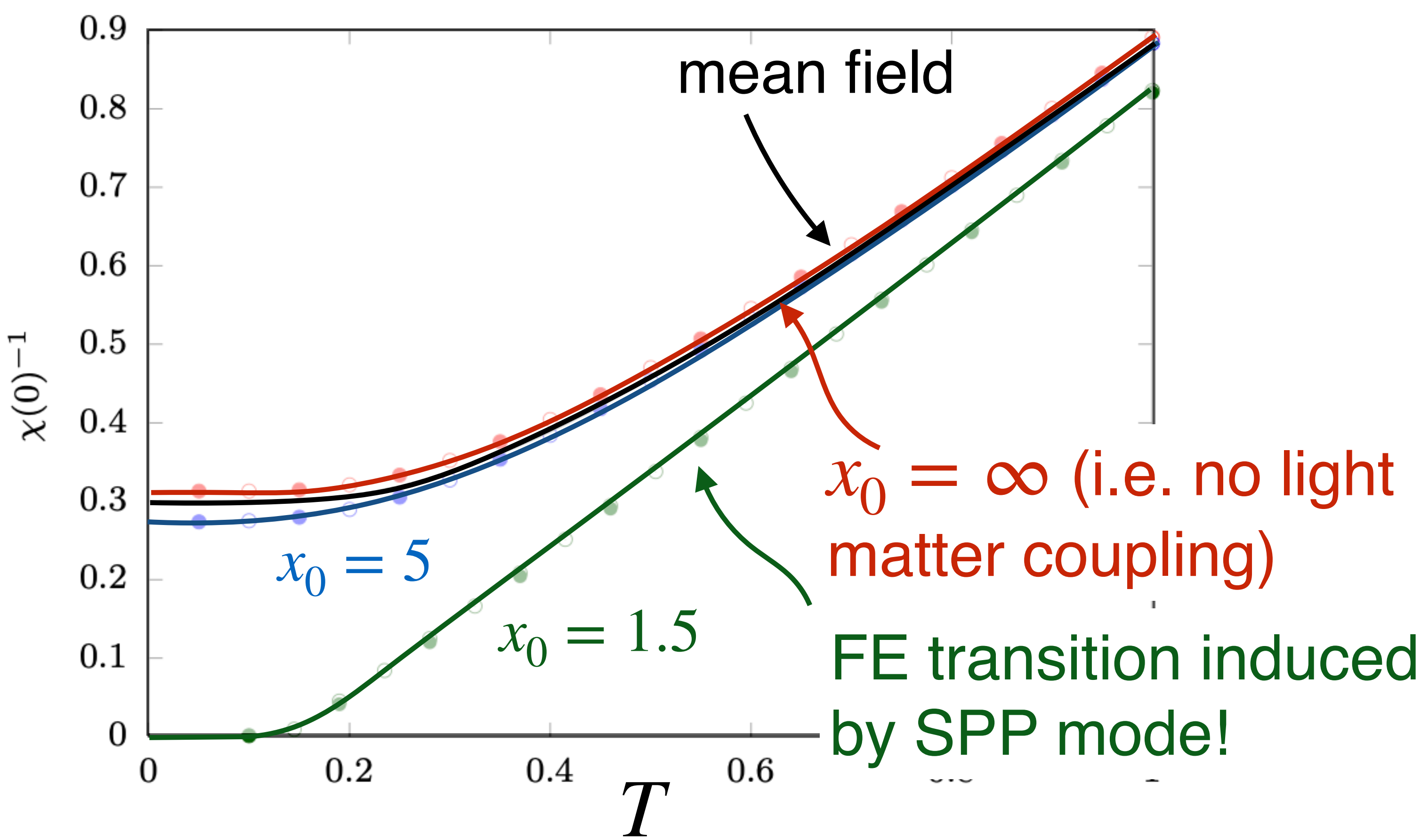
⇨ Interaction (in $q_{||}$) controlled by x_0

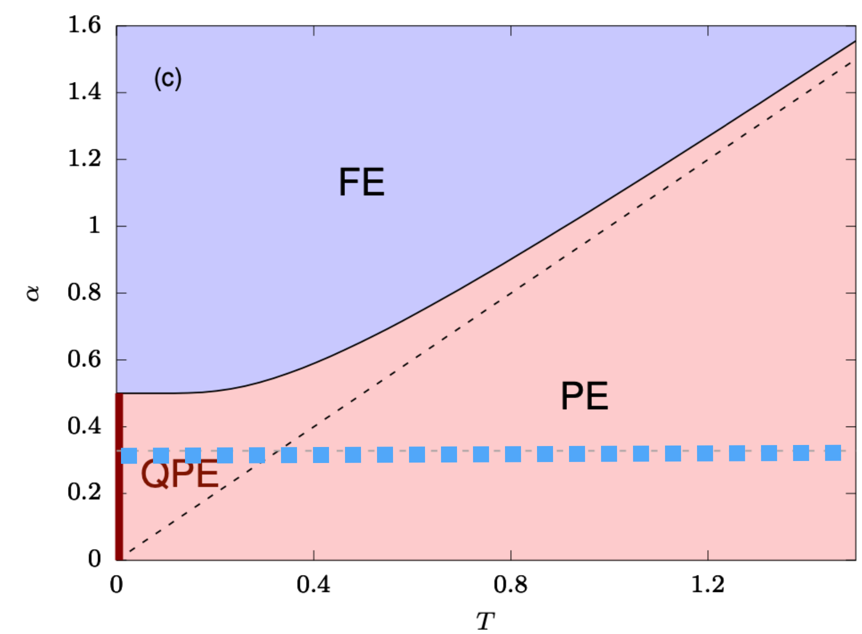




Back to “STO model”

Ferroelectric susceptibility $(T > T_c)$





Part 2: Experiment — *1T*-TaS_2 in a tunable cavity

Giacomo Jarc, S Mathengattil, A Montanaro, F Giusti, E Rigoni, F Fassioli, S Winnerl, S Dal Zilio, D Mihailovic, P Prelovšek, ME, Daniele Fausti, arXiv:2210.02346

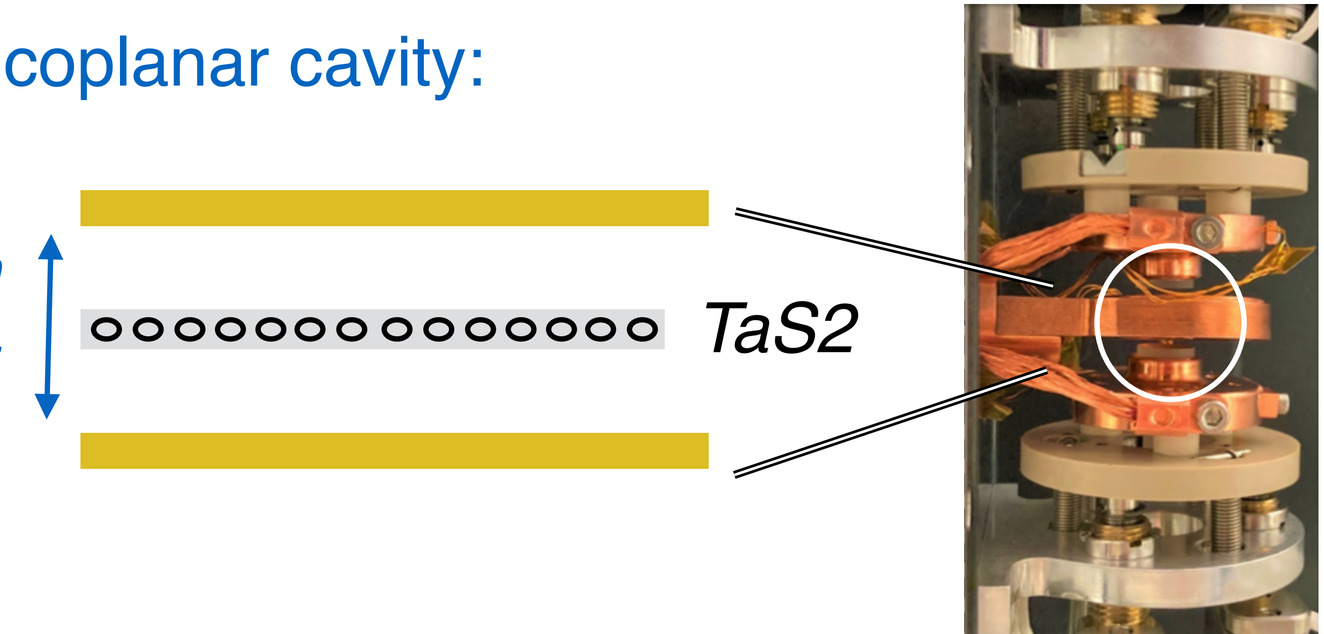
TaS2 in a tunable cavity

arXiv:2210.02346

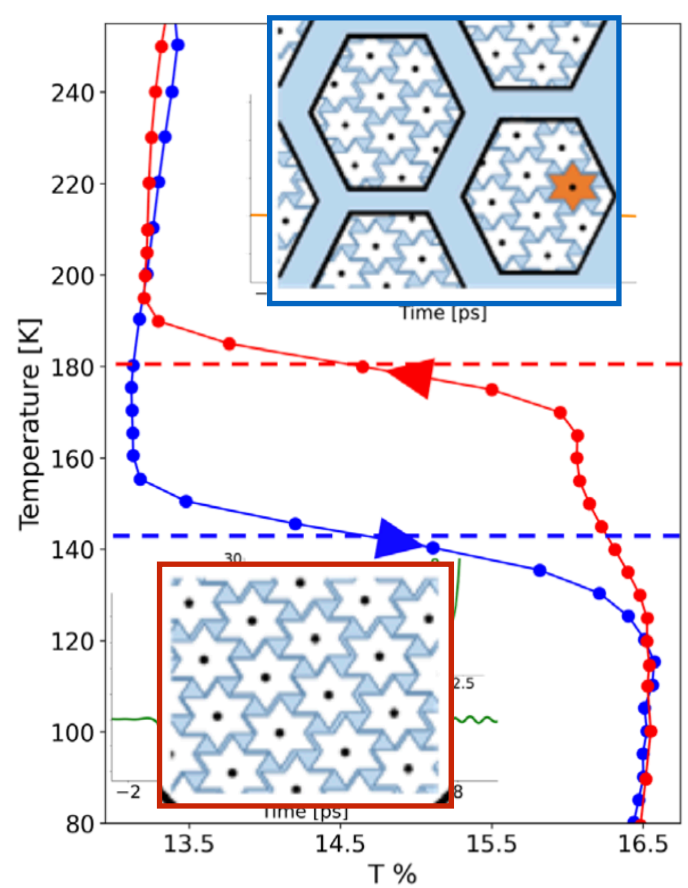
Tunable, optically accessible, cryogenic, coplanar cavity:

mm … cm

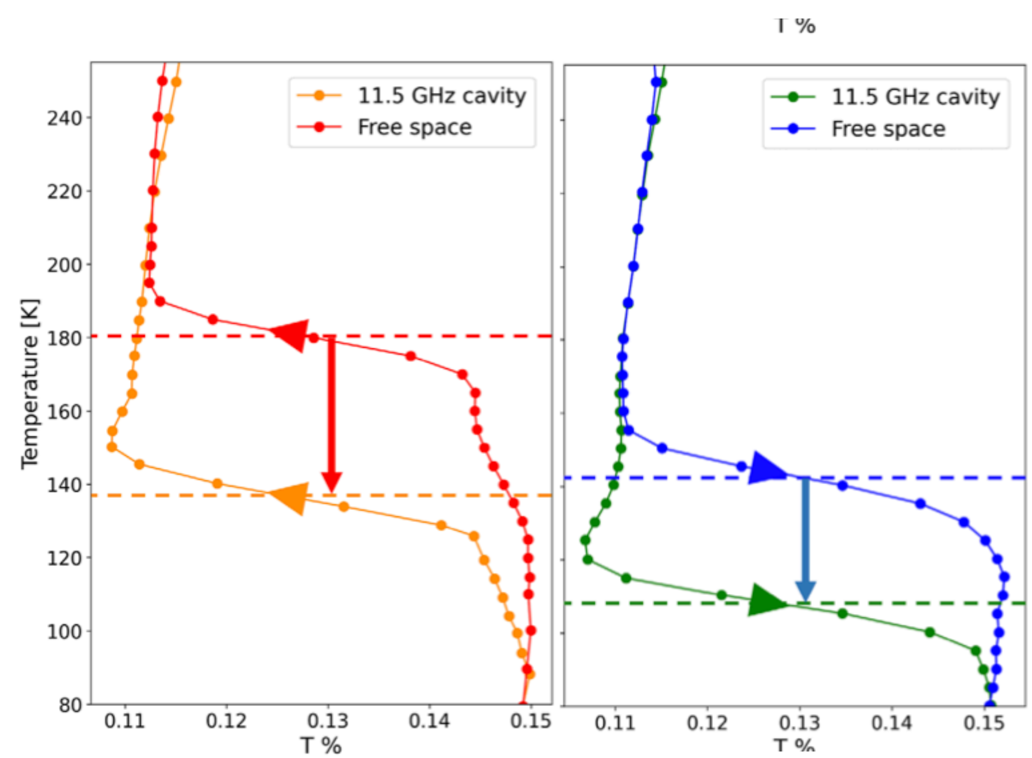
$\Omega = GHz \ldots THz$



TaS2: (optically detectable) 1st order metal insulator transition:



some optical signature sensitive to transition

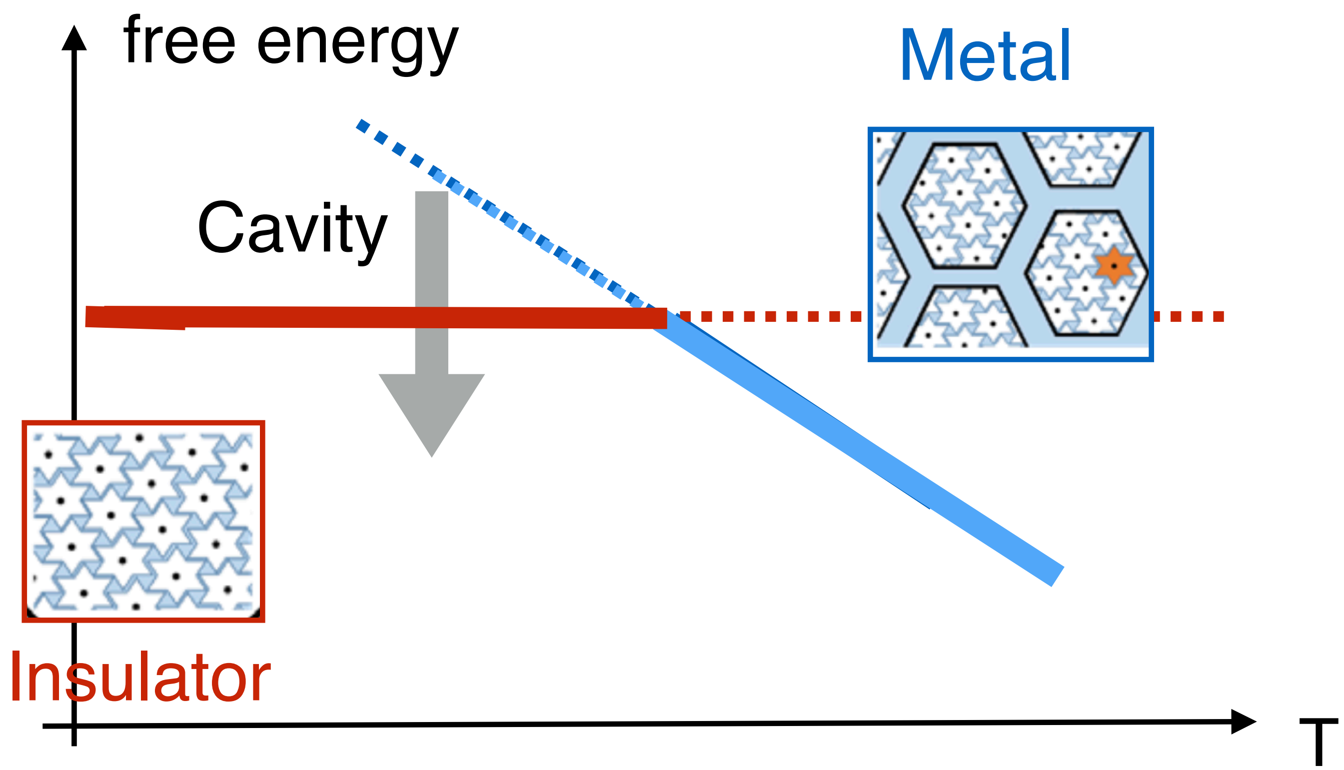


Transition in cavity shifted by up to 70K!

TaS2 in a tunable cavity

arXiv:2210.02346

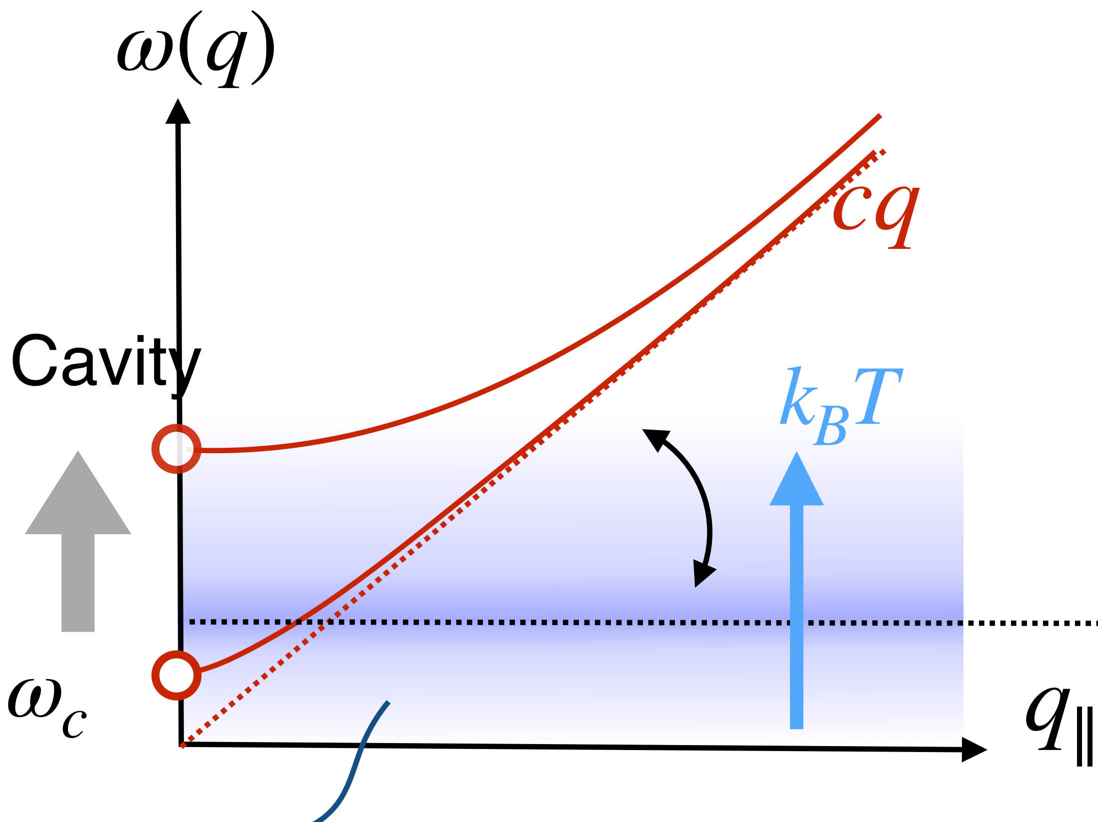
Possible explanation (1): “Selective Casimir effect”?



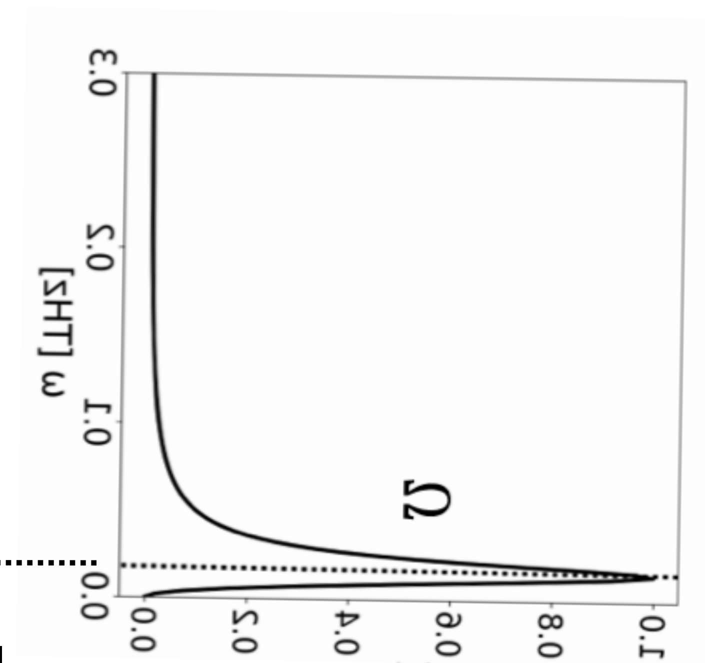
Selective change of free energy in metal due to formation of light-matter hybrid state?

TaS_2 in a tunable cavity

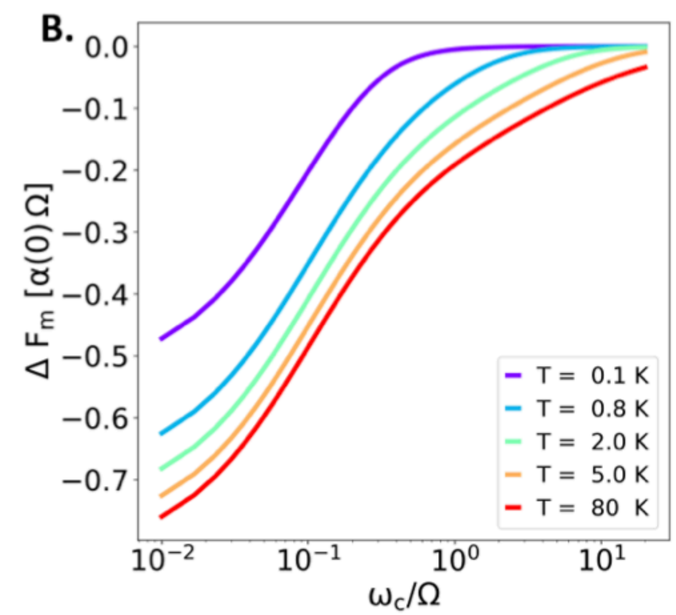
Change of free energy due to formation of light-matter hybrid state?



Electromagnetically active modes at Ω



Model dielectric loss $\alpha''(\omega)/\alpha'(0)$ at $q = 0$



ΔF (per mode):

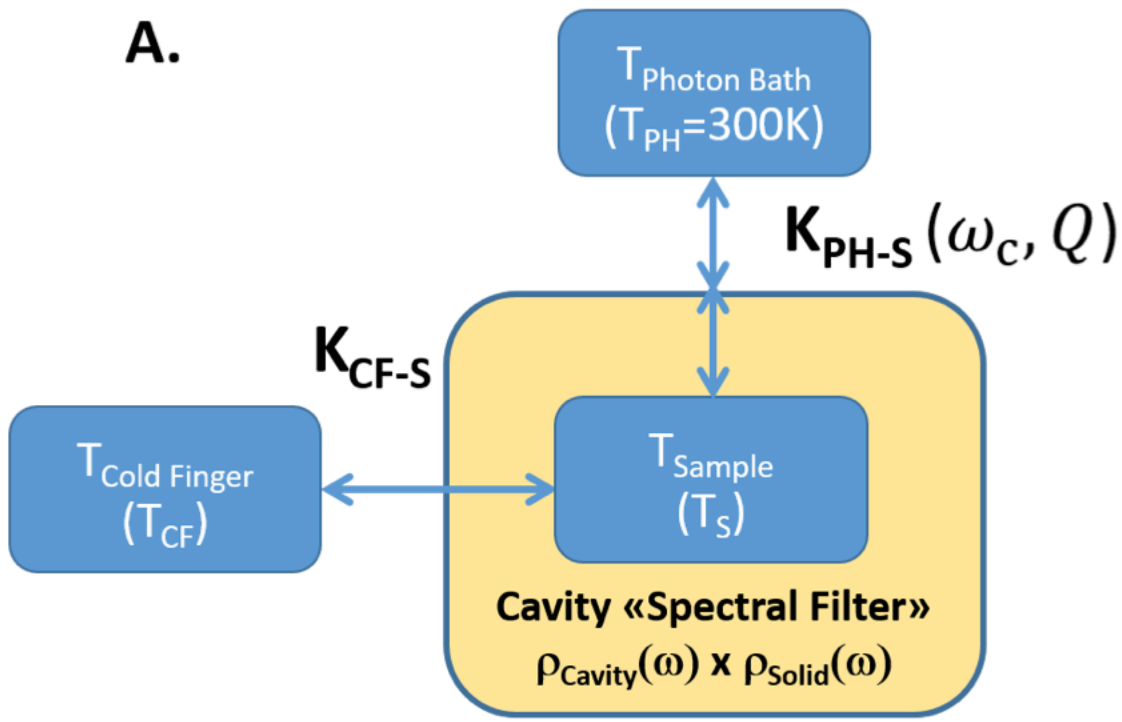
Estimate based on linear hybridization

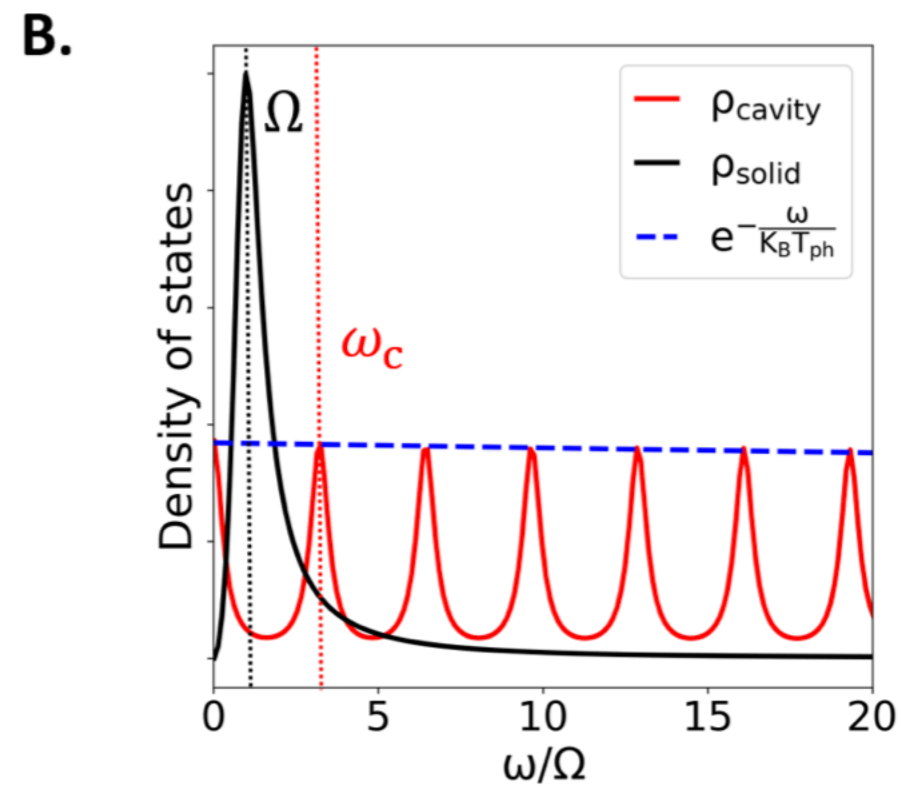
Phase space restriction $\rightarrow$ effect expected small for present setting

Generic effect of cavity on first order phase transition: ⇨ Again more relevant for plasmonic control of phase transition?

TaS_2 in a tunable cavity

Possible explanation (2): “Selective Purcell effect”?



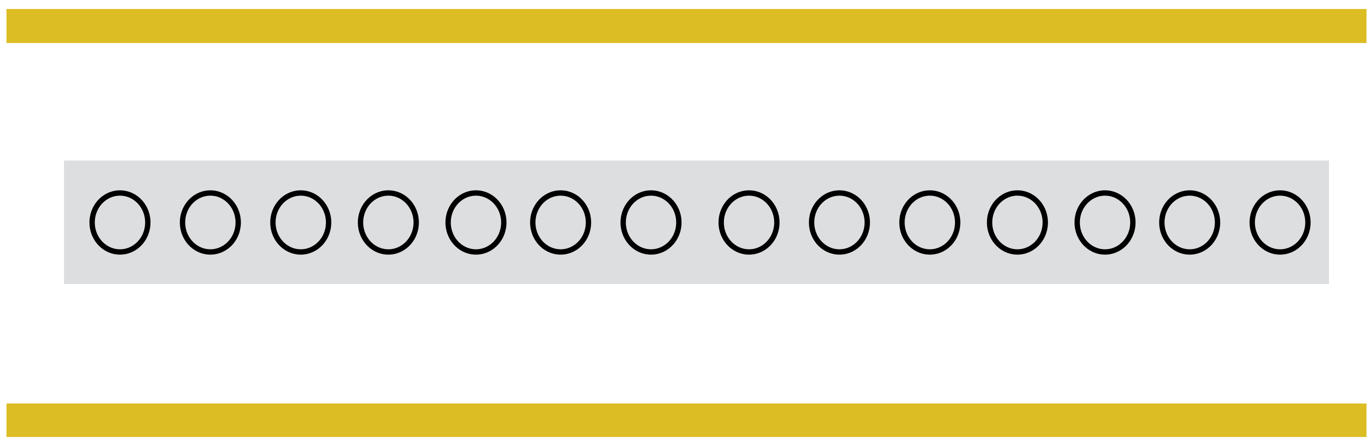


Cavity

⇨ redistribution of electromagnetic density of states

⇨ selective decoupling of fluctuations which are relevant for phase transition, and cavity (nonequilibrium steady state)

Summary



Engineer coupling of an extended solid to the *vacuum fluctuations* of the electromagnetic field to manipulate ground state and thermodynamic properties?

- Coupling to transverse field can help to condense ferroelectric modes …

  Main problem: Efficient coupling to continuum of modes?

  Here: Controlling matter by Couling to SPP mode!

  → Extensions to include also itinerant electrons, other order parameters, first order transition

- Experiment: Cavity-induced 1st order metal-insulator transition.

  → using Purcell effect to control state of system?

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Ferroelectric transition

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F Giusti
E Rigoni
S Winnerl
S Dal Zilio
D Mihailovic Ljubljana
P Prelovšek

DFG Deutsche Forschungsgemeinschaft

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