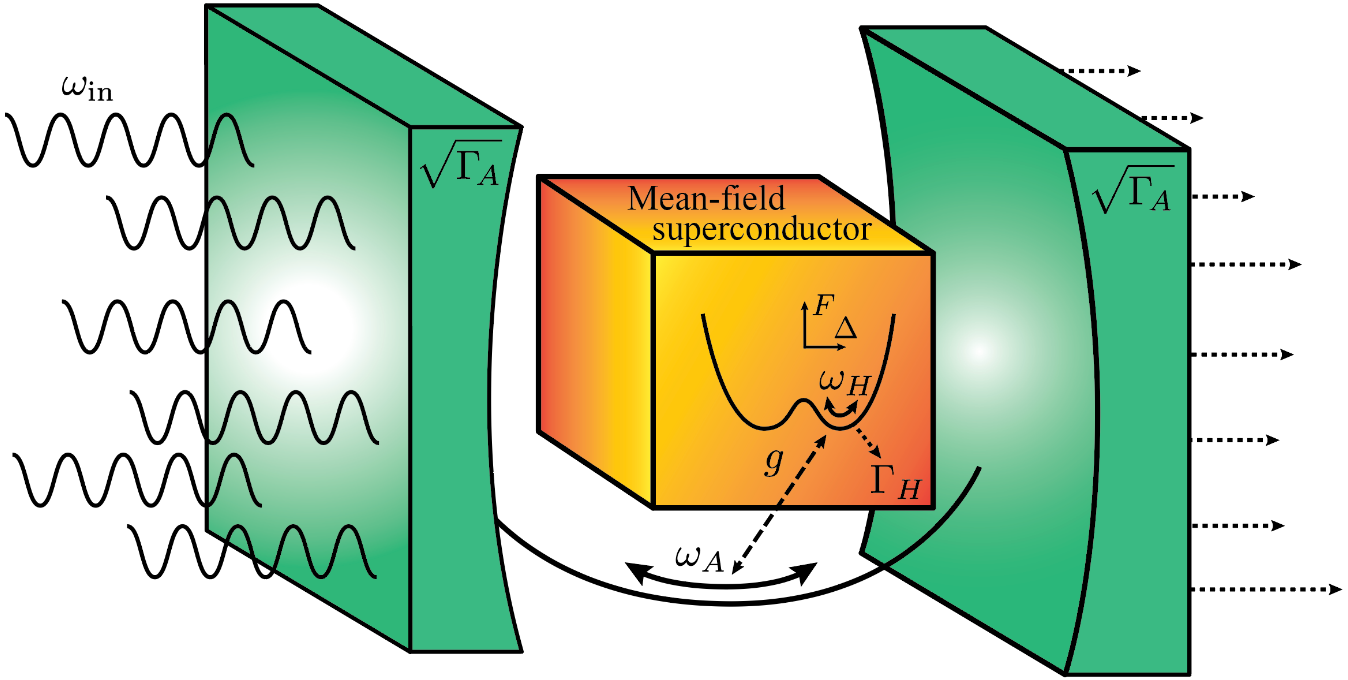
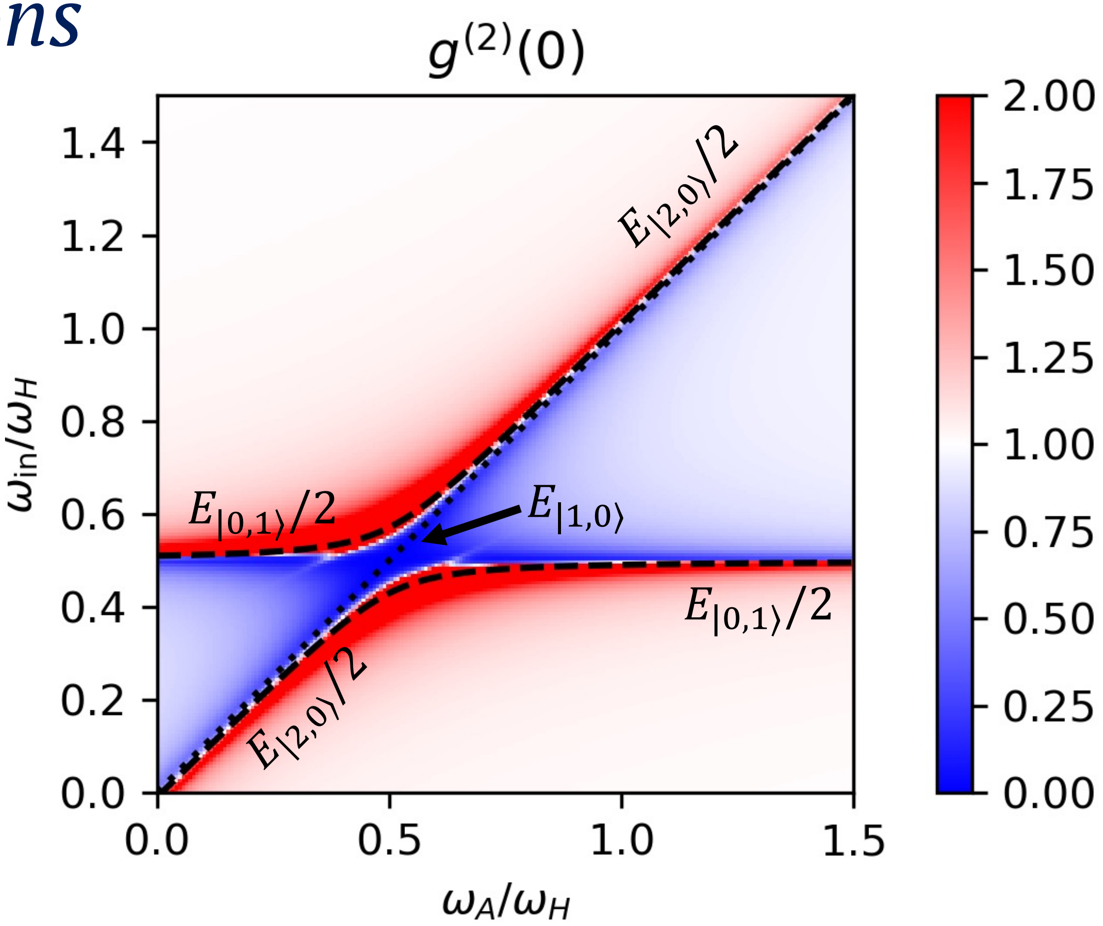
Cavity quantum material correlations from Keldysh field theory: *Photon Blockade from Higgs Polaritons*

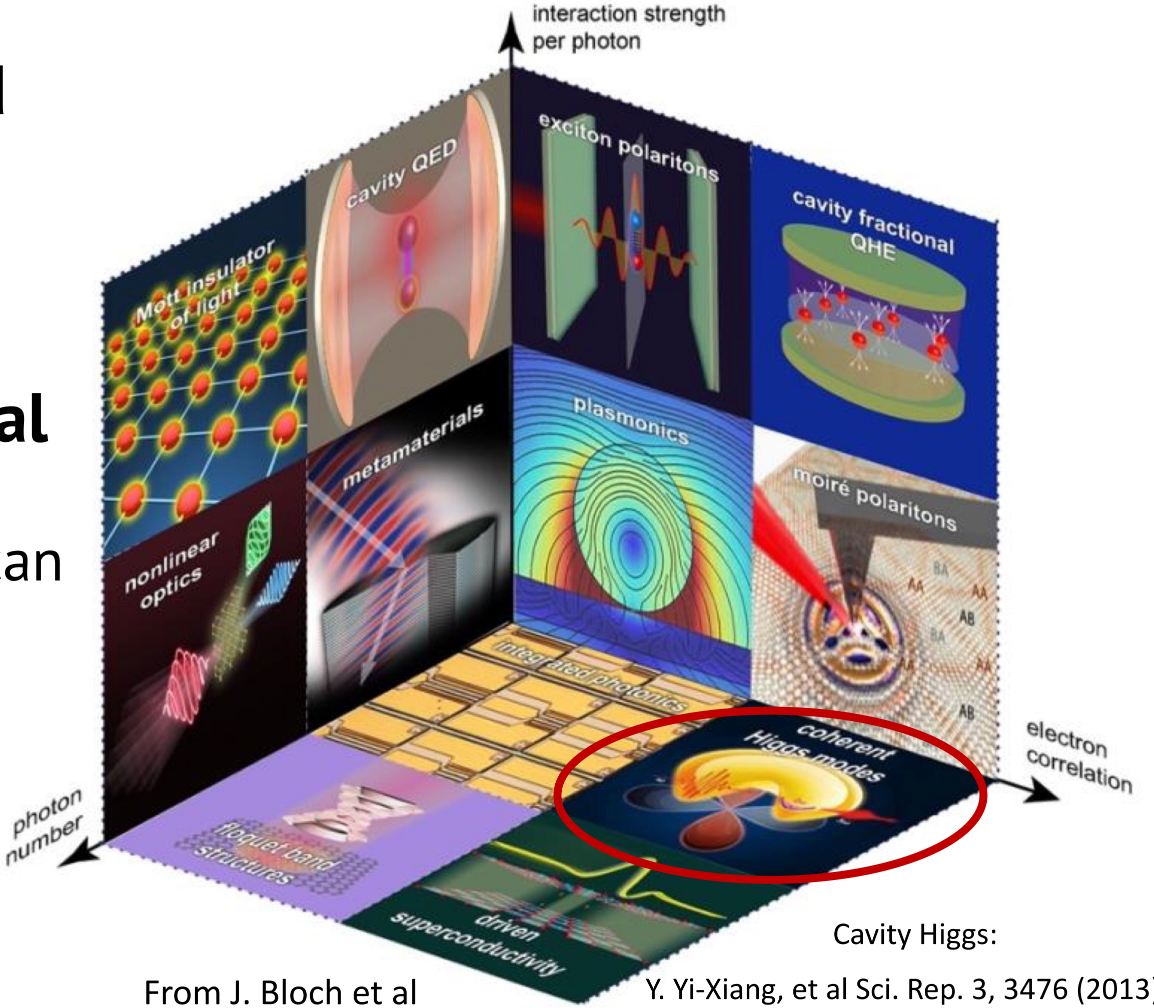
Spenser Talkington, Benjamin Kass, Martin Claassen & Ajit Srivastava • 20 March 2025





Cavity Quantum Materials

- To modify material properties, we need strong light-matter coupling
  - High fluence pulses (transient)
  - Low fluence cavities (steady state)
- **Alternative idea: what does the material do to the light? Acts as a nonlinearity**
  - With a collective order (e.g. a superfluid) can behave as an extended single emitter
  - Cavity materials can generate single THz photons from coherent driving
- Why do we want single THz photons?
  - Sensitive probes of *other* materials
  - Information processing



From J. Bloch et al Nature 606, 41 (2022)

Cavity Higgs:

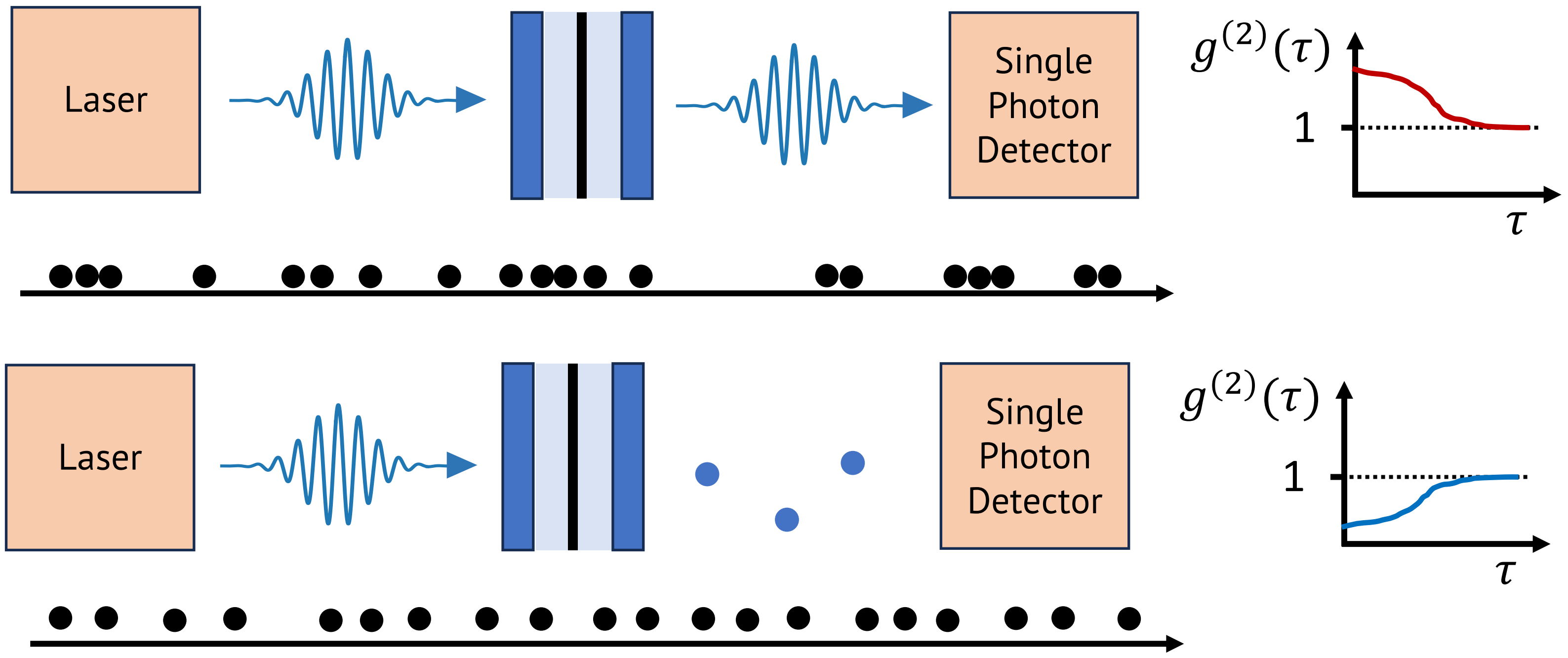
Y. Yi-Xiang, et al Sci. Rep. 3, 3476 (2013)
R. Matsunaga, Science 345, 1145 (2014)
Z.M. Raines, et al PRR 2, 013143 (2020)
H. Gao, et al PRB 104, 140503 (2021)

Transmission and Photon Counting

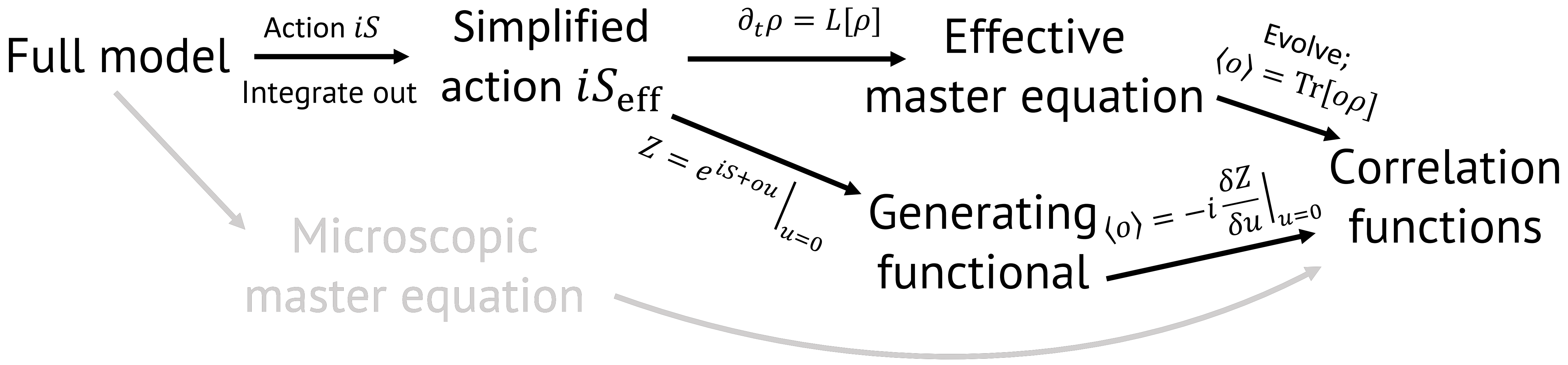
- Transmission: all light passed through the cavity—no back-scattering
- Photon (anti)-bunching from $g^{(2)}(\tau) = \frac{\langle a^{+}(t)a^{+}(t+\tau)a(t+\tau)a(t)\rangle}{\langle a^{+}(t+\tau)a(t+\tau)\rangle\langle a^{+}(t)a(t)\rangle}$
- If $g^{(2)}(0) \to 0$, only single photons are emitted (from a coherent drive)

Bunching
$g^{(2)}(0) > 1$

Anti-bunching
$g^{(2)}(0) < 1$



Field Theory for Constructing Models

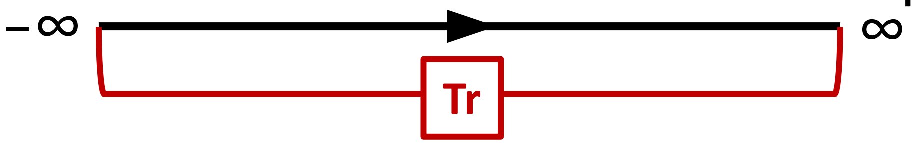


- What do we gain with a field-theoretic approach?
  - Compact description of low energy collective modes
  - Generation of models by integrating out / effective action
  - Rigorous perturbation theory in powers of many-body operators
  - Treat non-thermal or entangled input sources of light

Non-Equilibrium Path Integral

- A very useful object is the partition function Z = Tr[ρ] = 1
  - Equilibrium—final state is the initial state up to a phase

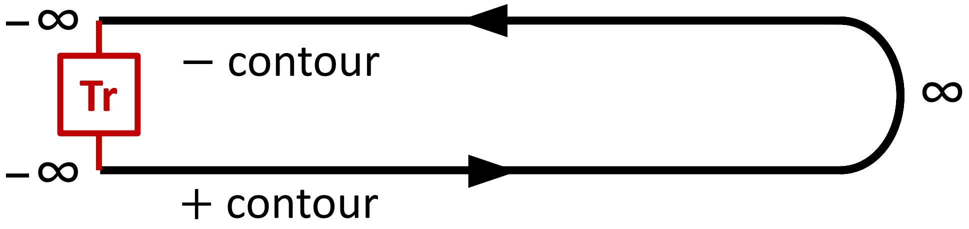
Z =



$$\psi(t) = U(t - t_i)\psi(t_i)$$

  - Non-equilibrium (“Keldysh” contour)—final state unrelated to initial state

Z =



$$\rho(t) = U(t - t_i)\rho(t_i)U(t_i - t)$$

- Express Z=e^{iS} in terms of an action iS

$$iS[a^+, a^-] = \ln(\rho(-\infty)) \; + \int_{-\infty}^{\infty} dt \; [\bar{a}^+\partial_t a^+ - \bar{a}^-\partial_t a^- + \hat{\mathcal{L}}[a^+, a^-]]$$

- To convert to master equation $\partial_t \rho = \mathcal{L}[\rho]$: a^+ goes left, a^- right of ρ

Empty Cavity Input-Output Action

start with discrete Keldysh action

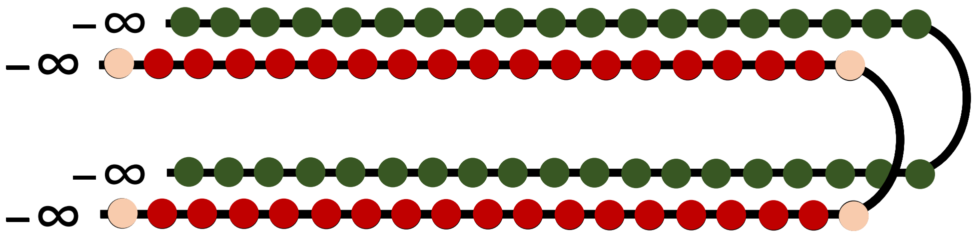
$$S[\{a_n\},\{b_n\}]$$

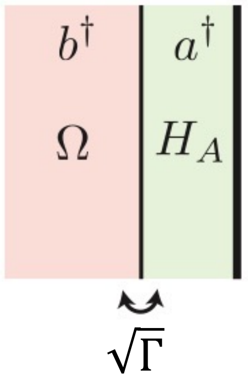
$\downarrow$ integrate out b bulk

$$S[\{a_n\}, b_1, b_N]$$

$\downarrow$ take continuum limit

$S[a, b_{\text{in}}, b_{\text{out}}]$ "Daniel-Potts action"





$$\begin{cases} iS[\boldsymbol{a},\boldsymbol{b}] = iS_A[\boldsymbol{a}] + iS_B[\boldsymbol{b}] + iS_{AB}[\boldsymbol{a},\boldsymbol{b}] \\ iS_B[\boldsymbol{b}] = \sum_\Omega \sum_{i=2}^{2N} \delta t\left[-\frac{\bar{b}_{i,\Omega}b_{i,\Omega} - \bar{b}_{i,\Omega}b_{i-1,\Omega}}{\delta t} - i\Omega\,\bar{b}_{i,\Omega}b_{i-1,\Omega}\right] \\ iS_{AB}[\boldsymbol{a},\boldsymbol{b}] = \sum_\Omega \sum_{i=2}^{2N} -i\sqrt{\Gamma}\delta t(\bar{a}_i b_{i-1,\Omega} + \bar{b}_{i,\Omega}a_{i-1}) \end{cases}$$

Action for T=0 coherent state $|f\rangle$ input, e.g. $f = f_0 e^{-i\omega_{\text{in}}t}$

$$iS_{\text{empty}}[a,f] = \int dt \begin{pmatrix}\bar{a}^+ \\ \bar{a}^-\end{pmatrix} \begin{pmatrix}\partial_t - iH_A - \Gamma & 0 \\ 2\Gamma & -\partial_t + iH_A - \Gamma\end{pmatrix} \begin{pmatrix}a^+ \\ a^-\end{pmatrix} + \int dt\, \sqrt{\Gamma}[f(\bar{a}^+ - \bar{a}^-) + \bar{f}(a^+ - a^-)]$$

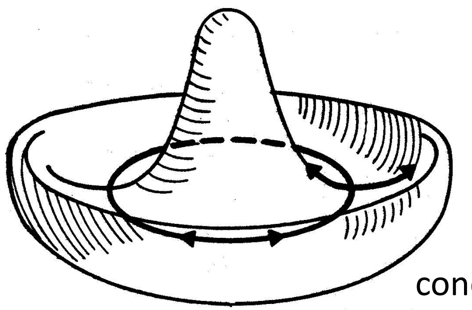
$$\partial_t \rho = \mathcal{L}_{\text{empty}}[\rho] = -i[H_A a^\dagger a + \sqrt{\Gamma}(f a^\dagger + \bar{f}a), \rho] - \Gamma a^\dagger a \rho - \Gamma \rho a^\dagger a + 2\Gamma a \rho a^\dagger$$

Higgs Modes In Superconductors

- Consider a mean-field superconductor with

$$F = F_0 - \alpha|\Delta|^2 + \beta|\Delta|^4 + g^\mu|(i\nabla^\mu + eA^\mu)\Delta|^2 \quad \boldsymbol{g} = (-P, g_0, g_0, g_0)$$

Ginzburg-Landau Free Energy

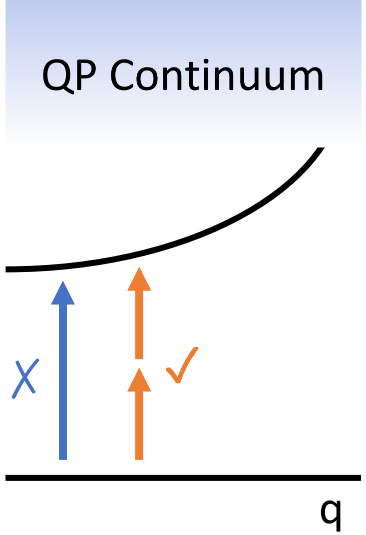


From C. Varma cond-mat/0109409

- At $T \ll T_c$ can expand in amplitude (Higgs) h and phase fluctuations θ: $\Delta = (\Delta_0 + h)e^{i\theta}$

$$F = h(P\partial_{tt} + 2\alpha)h + g_0 e^2 A^2(\Delta_0^2 + 2\Delta_0 h)$$

- With no damping, amplitude / phase modes decouple
- Stationarity of F gives $\omega_H = \sqrt{2\alpha/P}$, and $F' = \omega_H hh + gA^2 h$ where we discarded the shift and introduced $g = 2\Delta_0 e^2 g_0$
- F has $A^2 h$ term, but no Ah term (Higgs is neutral)
  - Two photon resonance can couple to Higgs
  - One photon resonance/linear response is forbidden



Master Equation Formulation

- We recall that $(f = f_0 e^{-i\omega_{\text{in}} t})$

$$\mathcal{L}_{\text{empty}}[\rho] = -i[\omega_A a^\dagger a + \sqrt{\Gamma_A}(f a^\dagger + \bar{f} a), \rho] + \Gamma_A(a^\dagger a \rho + \rho a^\dagger a - 2a\rho a^\dagger)$$

- Including the material we have

$$S_{\text{empty}} = S(H_A) \to S_{\text{full}} = S(H_A + F)$$

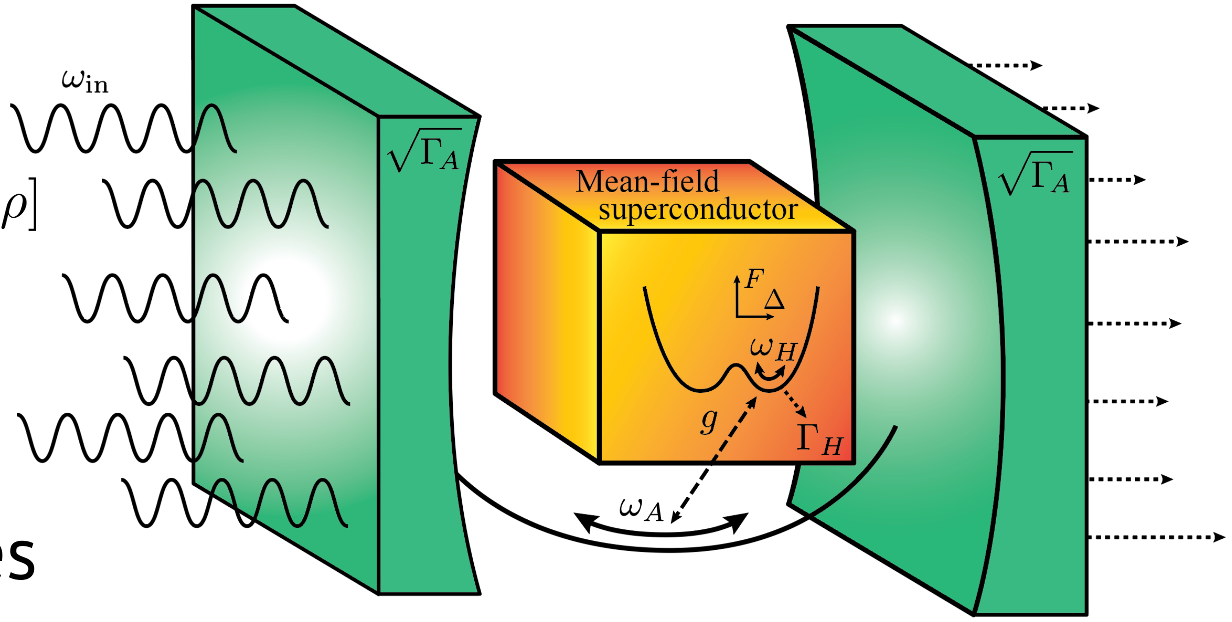
- And the master equation becomes

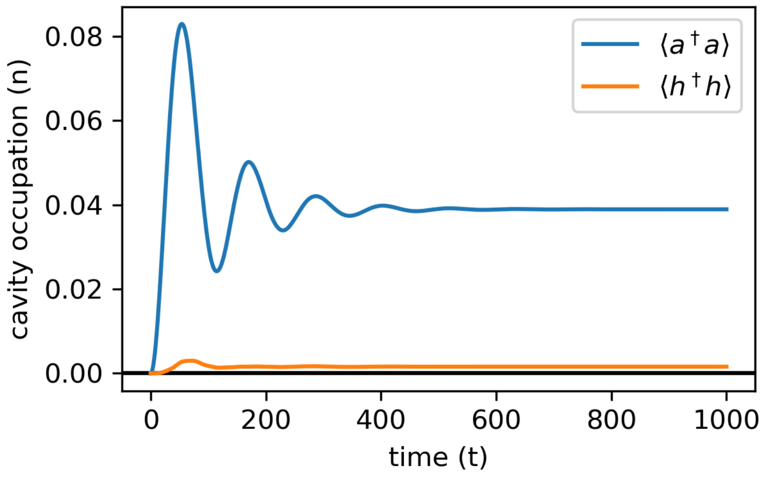
$$\partial_t \rho = (\mathcal{L}_{\text{empty}} + \mathcal{L}_F)[\rho]$$

- And now we have

$$\mathcal{L}_F[\rho] = -i[\omega_H h^\dagger h + g(aah^\dagger + a^\dagger a^\dagger h), \rho] - \Gamma_H(h^\dagger h \rho + \rho h^\dagger h - 2h\rho h^\dagger)$$

- Numerically evolve and $\langle a^+ a \rangle = \mathrm{Tr}[a^+ a\, \rho]$

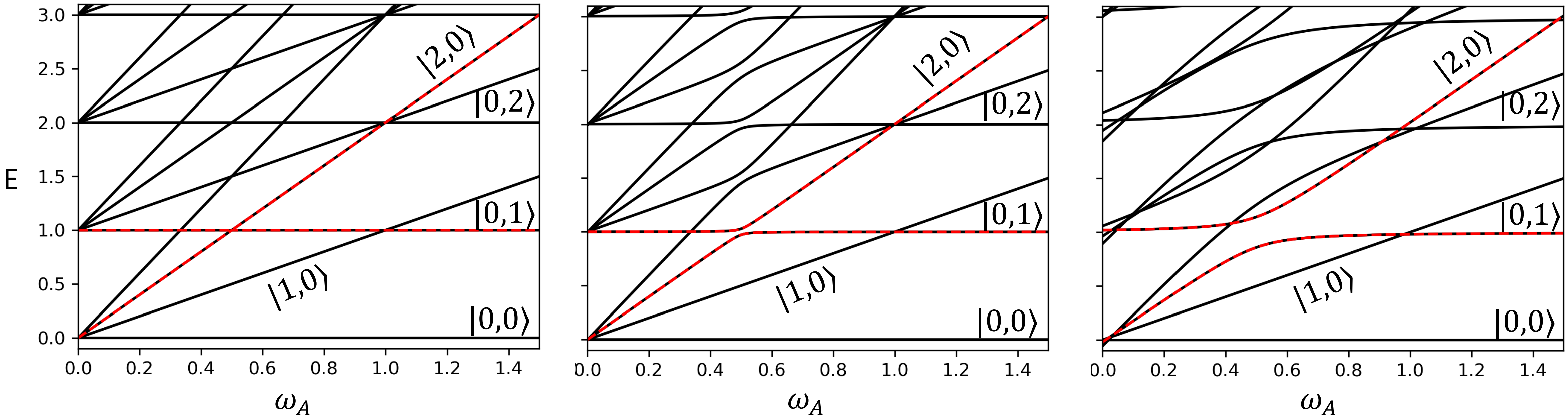




Higgs Polaritons

- Ignoring dissipation and driving $H = \omega_A a^+ a + \omega_H h^+ h + g(aah^+ + a^+ a^+ h)$
- States are polaritonic light-matter hybrids
- Fock states $|n_A, n_H\rangle$
- **Key idea: $E_{|2,0\rangle} \neq 2E_{|1,0\rangle}$**

$$H_{\text{red}} = \begin{pmatrix} \langle 2,0|H|2,0\rangle & \langle 2,0|H|0,1\rangle \\ \langle 0,1|H|2,0\rangle & \langle 0,1|H|0,1\rangle \end{pmatrix} = \begin{pmatrix} 2\omega_A & \sqrt{2}g \\ \sqrt{2}g & \omega_H \end{pmatrix}$$

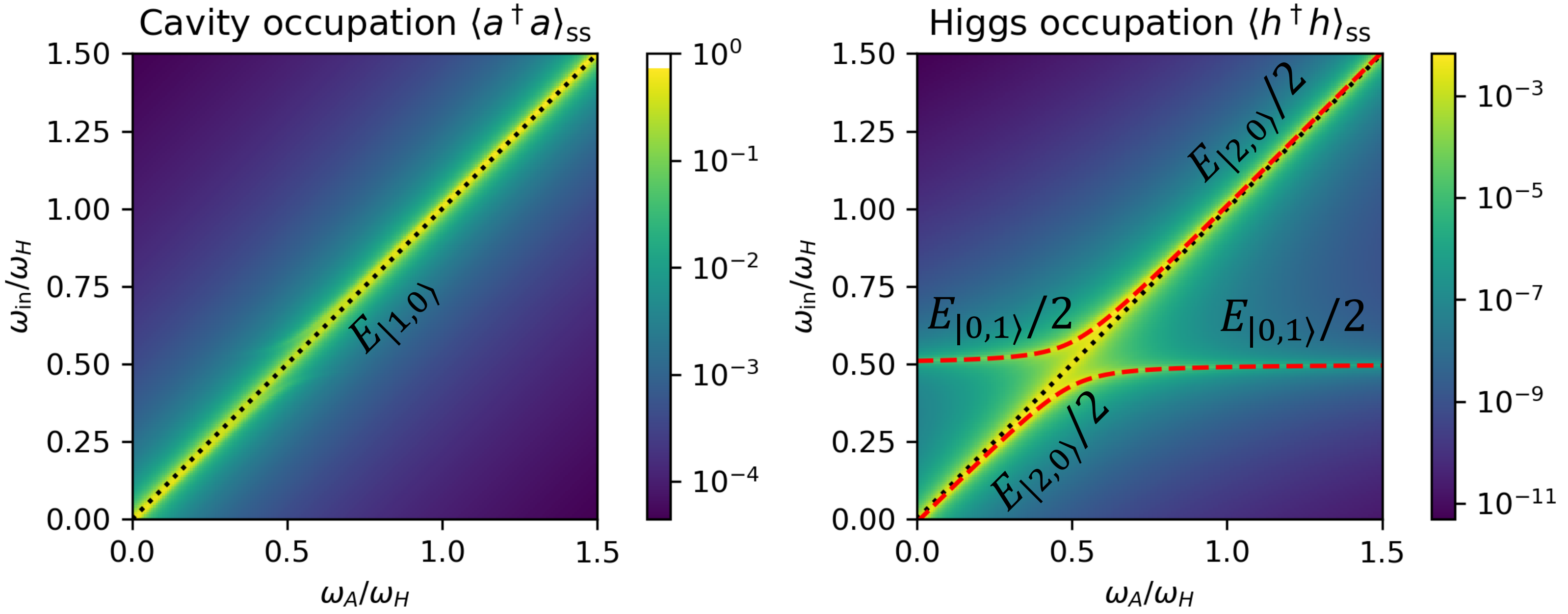


Light Emitted From Higgs + Cavity

- Density is given by resonant driving condition $\omega_{\text{in}} = \omega_A$
- Antibunching: single photon in Higgs polariton gap
- Bunching: Higgs decay into two cavity photons

3 Frequencies
- ω_H Higgs
- ω_A cavity
- ω_{in} drive

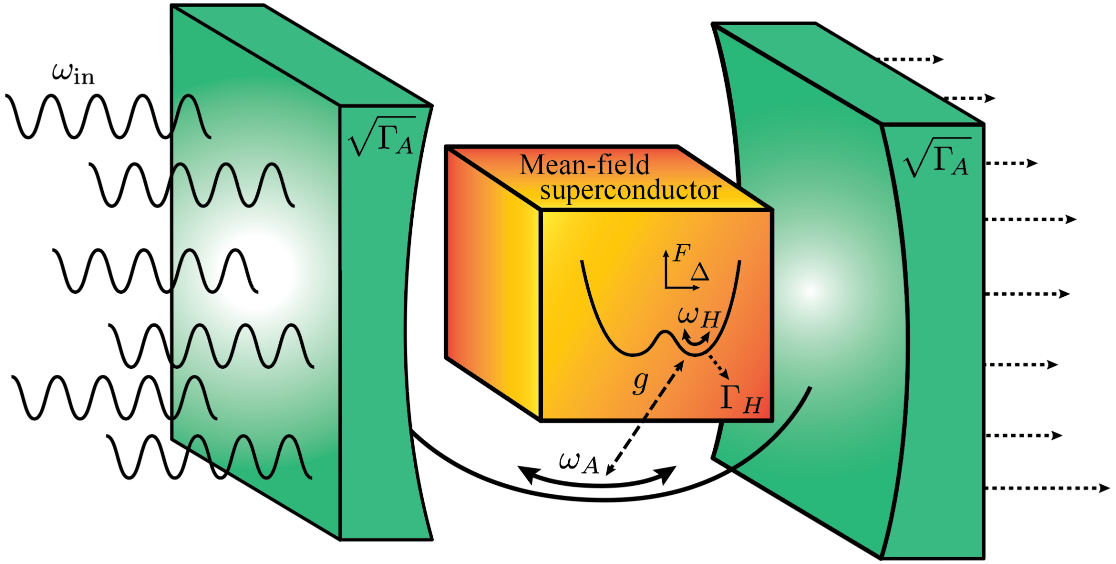
$M_{\max} = 4$
$\omega_H = 1$
$f = 0.1$
$\Gamma_A = 0.01$
$\Gamma_H = 0.01$
$g = 0.1$
$(Q = 50)$



Outlook

- Cavity quantum materials can both be probed by the output light and used as a resource to generate quantum light
- Key idea: to populate |2,0> state we need to drive at a different frequency than |1,0> state, not just drive harder!
- Keldysh field theory provides a natural framework to address these systems
- THz sources and tunable cavities exist (Fassioli et al, 2403.00851), and single photon detectors are being developed

- *Talk by Benjamin Kass in 20 mins: quantum critical materials in cavities (optical Raman)*



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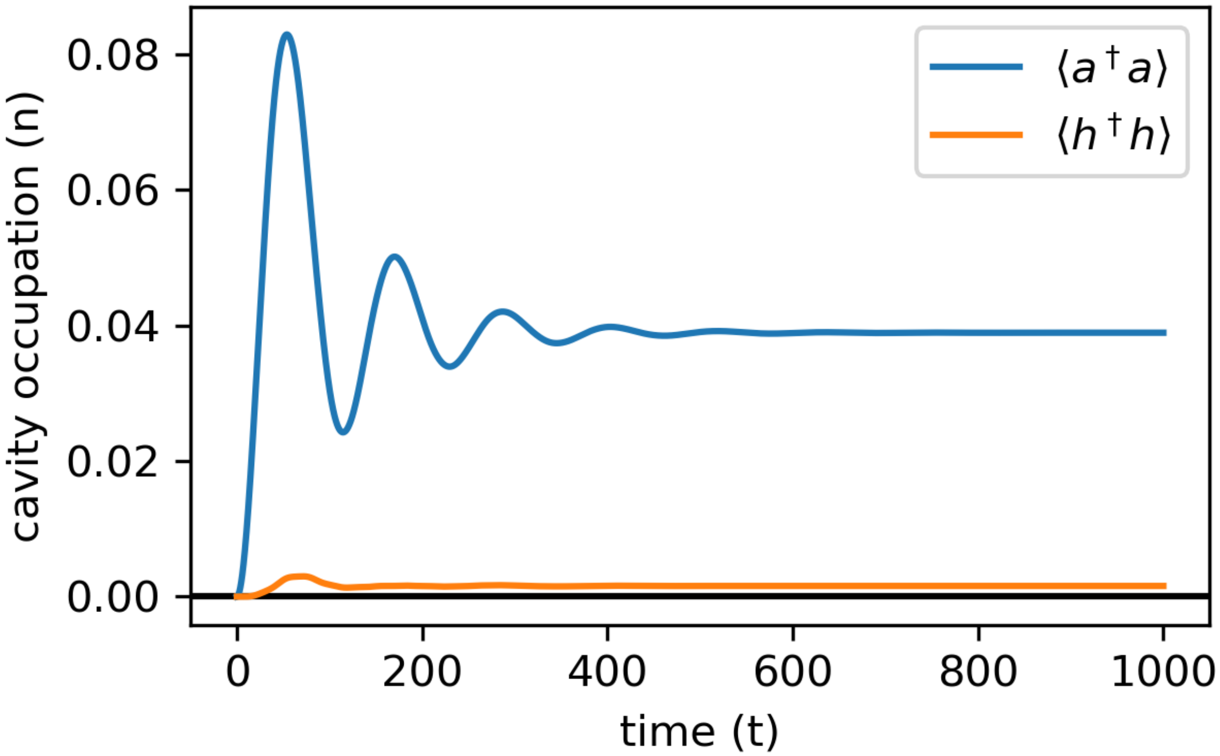
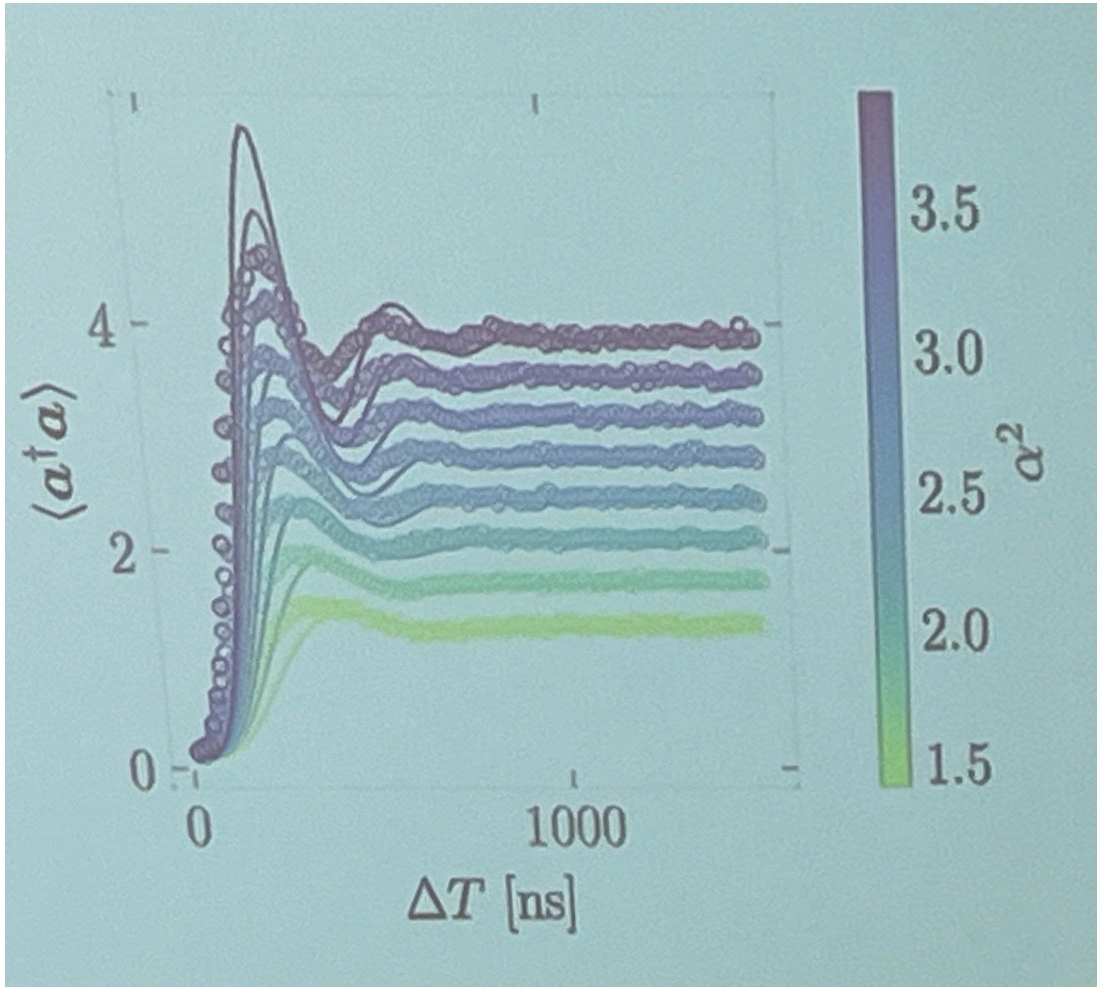
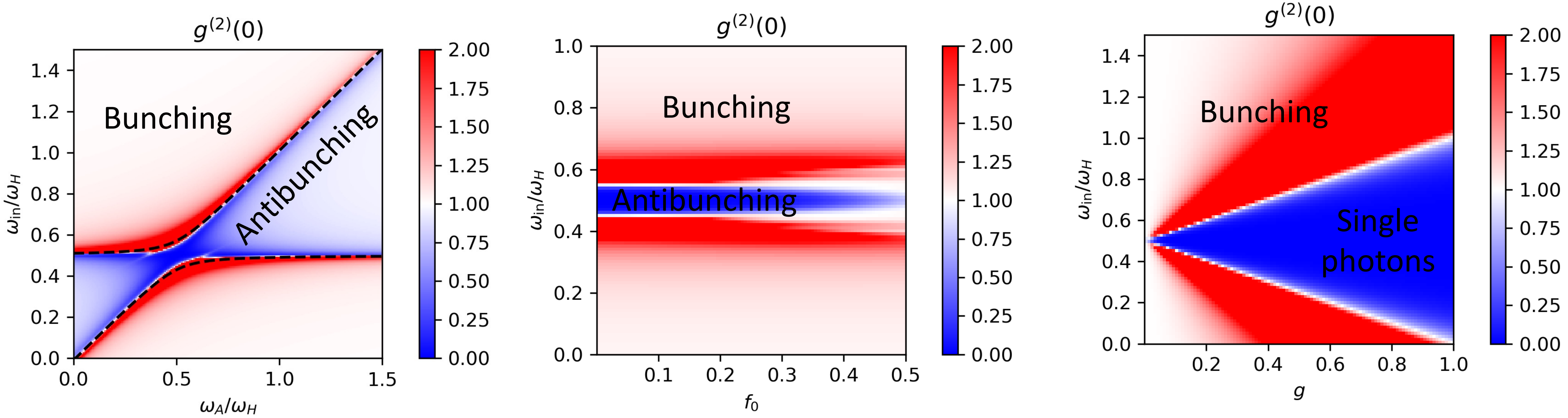




U. Reglade (Unpublished)

Strong Driving and Strong Coupling



$M_{\max} = 4$
$\omega_H = 1$
$\Gamma_a = 0.01$
$\Gamma_h = 0.01$
$(\omega_A = 1)$
$(f = 0.1)$
$(g = 0.1)$

EPR Pairs Emitted From Chiral Material

- Cavity embedded materials with spatial symmetries can be used to generate quantum entanglement
- E.g. EPR pairs from linearly polarized light: drive at ω_{in} to only address the entangled triplet eigenstate
- Preprint: 2411.09864
- *See talk by Benjamin Kass (in 24 minutes)*
  - *Quantum critical cavity embedded materials*
  - *Photon bunching and single photon emission*