Probing Quantum Materials with Quantum Light

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Abstract

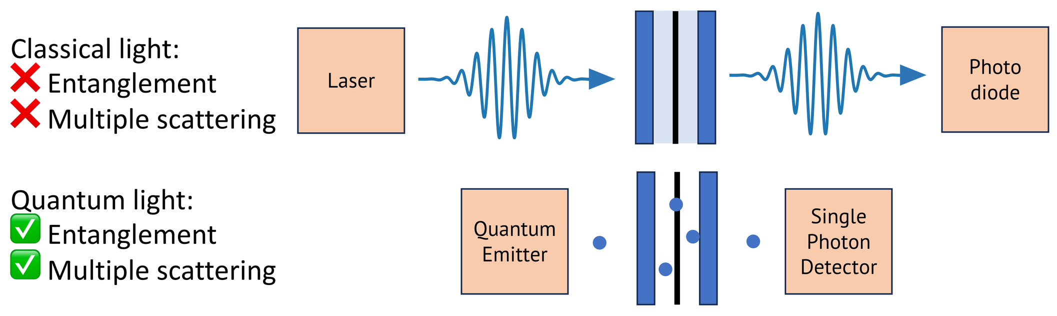
In recent years strong light-matter coupling in cavities has emerged as a promising mechanism to tune many-body correlations in electronic and spin systems. A key question remains in how to probe these states and show that they have the claimed ordering. Here we provide a formal treatment of the transmission of arbitrary multi-photon states through cavities, as a function of non-linear interactions with a quantum material. We do this by generalizing quantum optical input-output relations to extended quantum systems and introduce Keldysh input-output theory as a new, systematic way to calculate input-output correlation functions theory using Keldysh field theory.

Our Previous Works

Dissipation in quantum systems is not invariably inimical: it can be a resource to engineer desirable steady states. We have shown that dissipation can be used to engineer band structures [PRB 106, L161109 (2022)] and to realize non-equilibrium steady states and probe their dynamic response properties [arXiv:2401.17368, 2402.06593].

Input-Output Theory

Cavities provide a natural platform for the most exotic properties of quantum light such as entanglement and high-order correlations. Here we consider the response of cavity materials to arbitrary multiphoton states.



Cavity input-output theory as formulated by Gardiner and Collett [1] relates output light modes to the input modes and the features of the cavity via the relation

$$\hat{b}_{\text{out}}(t) = \hat{b}_{\text{in}}(t) + \gamma \hat{a}(t)$$

where γ quantifies the quality factor of the cavity.

Correlation Functions

The $g^{(1)}$ and $g^{(2)}$ functionals correspond to intensity and intensity fluctuations, respectively. The functionals are

$$g^{(1)}(t) = \langle a^\dagger(t) a(t) \rangle = \frac{i}{2}(G^K - G^R + G^A)$$

and

$$g^{(2)}(t,t') = \frac{\langle a^\dagger(t) a^\dagger(t+t') a(t+t') a(t) \rangle}{|\langle a^\dagger(t) a(t) \rangle|^2}$$

respectively where completing the convolutional time integral leads to δ(t-t') which creates loop diagrams. $g^{(2)}$ can also be expressed in terms of Green's functions.

$$G^<(t,t') = -i\langle a^\dagger(t') a(t) \rangle, \quad G^>(t,t') = -i\langle a(t) a^\dagger(t') \rangle$$

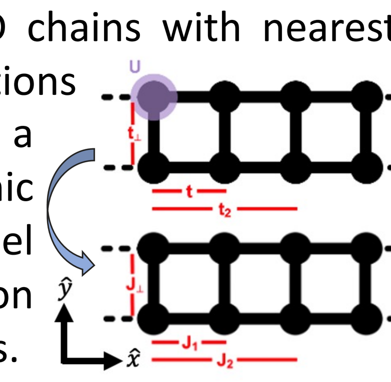
$$G^R = \theta(t-t')[G^> - G^<], \qquad G^K = G^> + G^<$$

where G^R tells us about the modes present and G^K tells us about the occupations. Note that $G^A = (G^R)^\dagger$

Example: J_1-J_2 Spin Ladder

System

As an example of the uses of input-output theory to predict the results of quantum photon spectroscopy of cavity quantum materials we present the example of a spin ladder with two coupled 1D chains with nearest and next-nearest neighbor interactions. This model could originate from a spin model or from an electronic model such as the Hubbard model at half filling where charge motion is frozen, but spins motion remains.

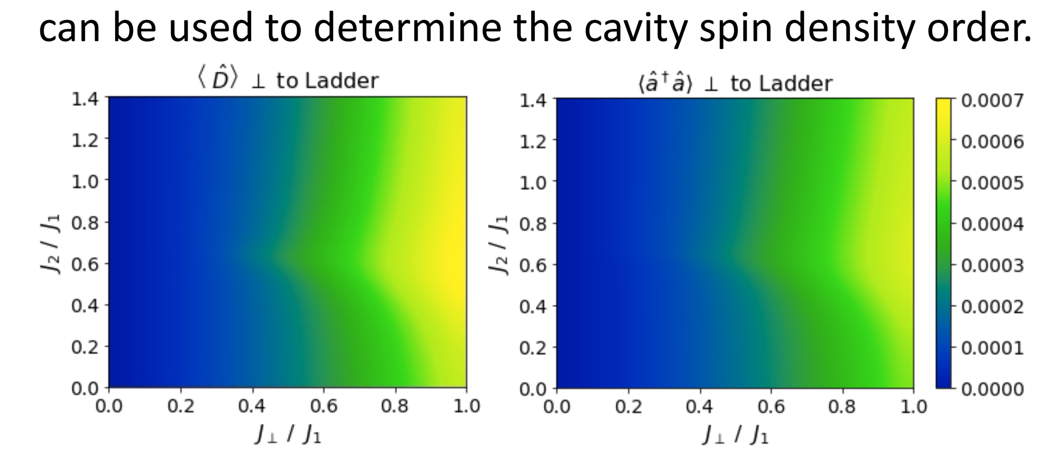


Cavity-Free Space Mode Correspondence

Using DMRG with Chebyshev polynomial expansion, Ben Kass found the material and output photon correlation functions. The $g^{(1)}$ function when reprocessed by

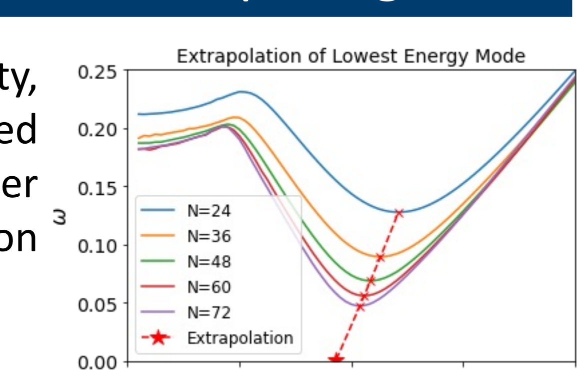
$$\hat{D} = \sum_{ij} C_{ij}(\hat{S}_i \cdot \hat{S}_j - \tfrac{1}{4})$$

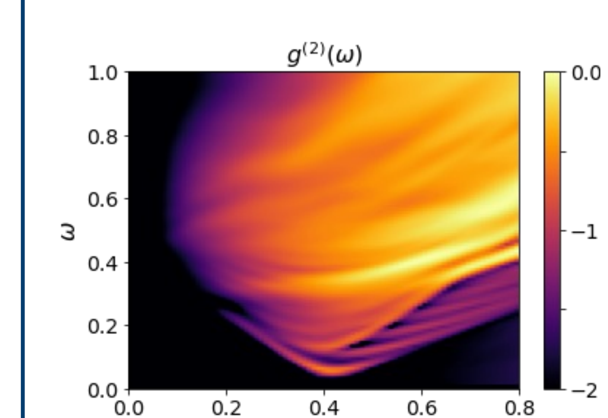
gives a precise quantitative agreement, so output light can be used to determine the cavity spin density order.

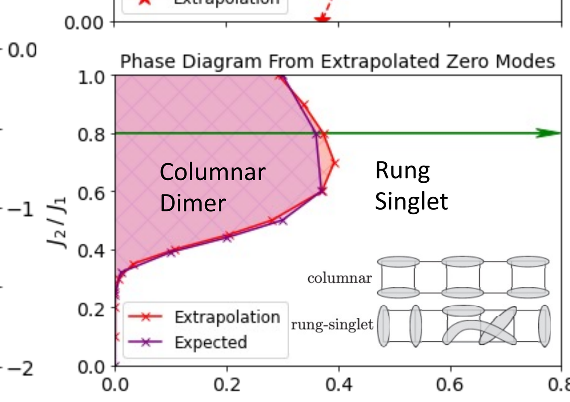


Phase Diagram from Output Light

In addition to the density, output light can be used determine order parameter from multipoint correlation functions such as $g^{(2)}$.

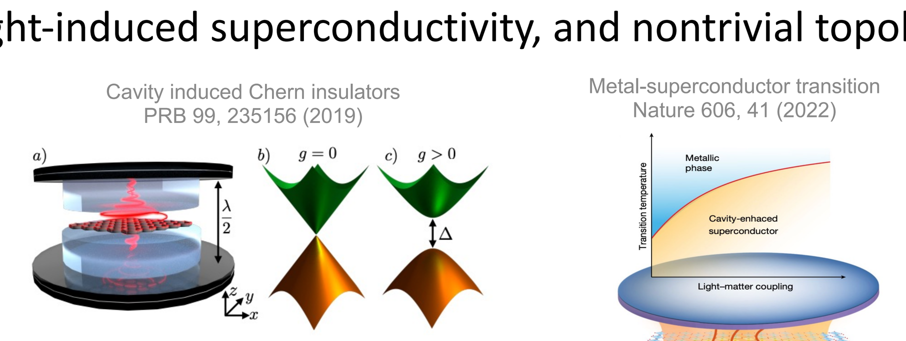




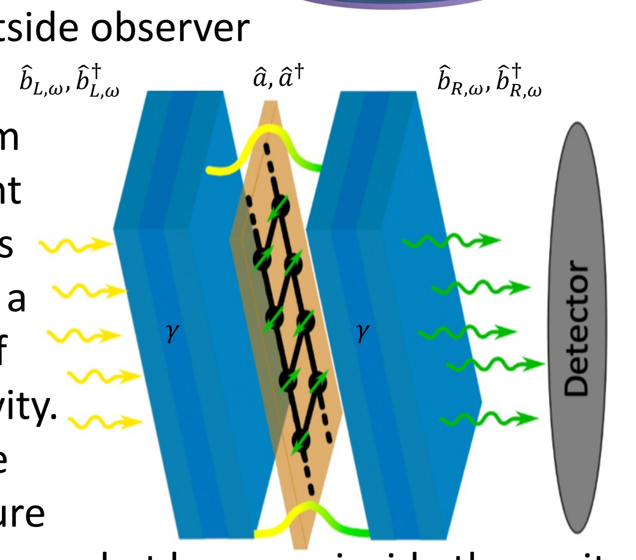


Cavity Quantum Materials

Cavities can facilitate strong light-matter coupling and hybrid light-matter states with distinct properties from the light or the matter alone such as polaritonic physics, light-induced superconductivity, and nontrivial topology [2]



But the physics in these cavities is inaccessible to an outside observer unless there is some input and output from the cavity such as light that enters and leaves the cavity because of a finite quality factor of the mirrors of the cavity. We can then seed the input light and measure the outgoing light to see what happens inside the cavity.



Field Theoretic Methods

Input-Output Continuum Action

We can write the continuum action of a bipartite system

$$iS[a, b_{\text{in}}, b_{\text{out}}, \psi] = iS_A + iS_B + iS_{AB} + iS_\psi$$

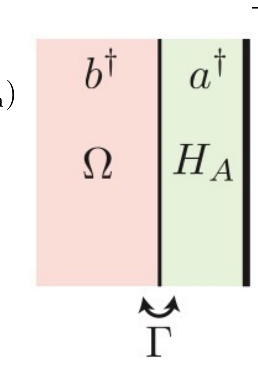
where the individual terms in the action are [4]

$$iS_A = i\int_{t_1}^{t_N} dt \left[\bar{a}^+\left(i\partial_t + i\frac{\Gamma}{2}\right)a^+ - \bar{a}^-\left(i\partial_t - i\frac{\Gamma}{2}\right)a^- - H_A(\bar{a}^+, a^+) + H_A(\bar{a}^-, a^-)\right]$$

$$iS_B = -[\int_{t_1}^{t_N} dt \begin{pmatrix} \bar{b}_{\text{in}}^+ \\ \bar{b}_{\text{out}}^+ \\ \bar{b}_{\text{out}}^- \\ \bar{b}_{\text{in}}^- \end{pmatrix} \begin{pmatrix} 1 & 0 & 0 & 0 \\ -1 & 1 & 0 & 0 \\ 0 & -1 & 1 & 0 \\ 0 & 0 & -1 & 1 \end{pmatrix} \begin{pmatrix} b_{\text{in}}^+ \\ b_{\text{out}}^+ \\ b_{\text{out}}^- \\ b_{\text{in}}^- \end{pmatrix}] + \ln(\rho_{\text{in}})$$

$$iS_{AB} = -\sqrt{\Gamma}\int_{t_1}^{t_N} dt\ \bar{a}^+ b_{\text{in}}^+ - \bar{b}_{\text{out}}^+ a^+ - \bar{a}^- b_{\text{out}}^- + \bar{b}_{\text{in}}^- a^-$$

$$iS_\psi = -i\int_{t_1}^{t_N} dt\ \bar{\psi} b_{\text{out}}^+ + \bar{b}_{\text{out}}^- \psi$$



and iS_ψ is added to generate moments at t_{out}.

Computational Steps

The key simplification that allows us to make progress is that while the material is strongly interacting, and there can be many-photon processes, the photons are non-interacting so we can integrate them out exactly.

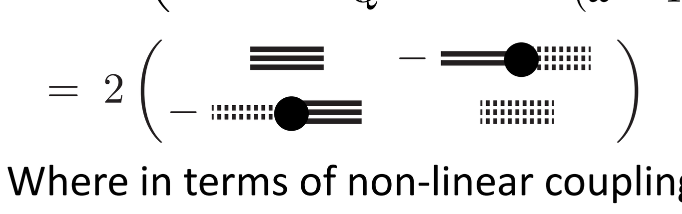
Once we have integrated out, we have (1) parameters for the system, (2) data about the initial state, and (3) many-body operators of the material. Together these parts manifest in the output correlation functions.

$S[a, b_{\text{in}}, b_{\text{out}}, \psi]$
$\downarrow$ integrate out b_{out}
$S[a, b_{\text{in}}, \psi]$
$\downarrow$ put in input state
$S[a, b_{\text{in}}, \psi]$
$\downarrow$ integrate out b_{in}
$S[a, \psi]$
$\downarrow$ Larkin rotate
$S[a, \psi] \to G^R,\ G^A,\ G^K$
$\downarrow$ integrate out a
$S[\psi] \to$ calculate moments

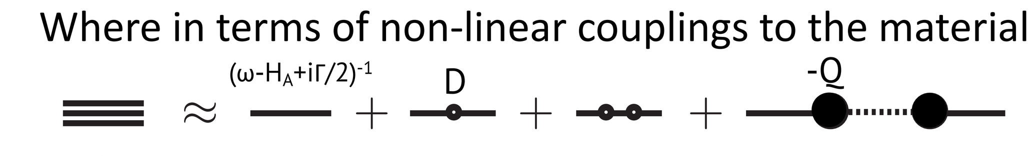
Green's Functions and Diagrammatics

The correlation functions we want are all expressed as convolutional integrals in terms of the Green's functions

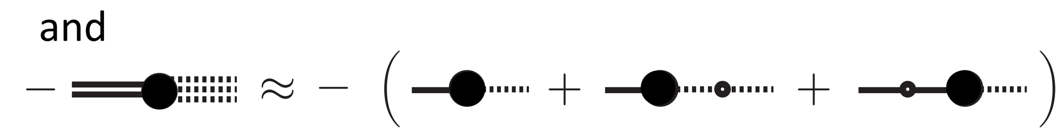
$$G^R = 2 \cdot \begin{pmatrix} \omega - H_A - \hat{D} + i\Gamma/2 & -\hat{Q} \\ -\hat{Q} & (\omega - H_A - \hat{D} + i\Gamma/2)^* \end{pmatrix}^{-1}$$



Where in terms of non-linear couplings to the material



and

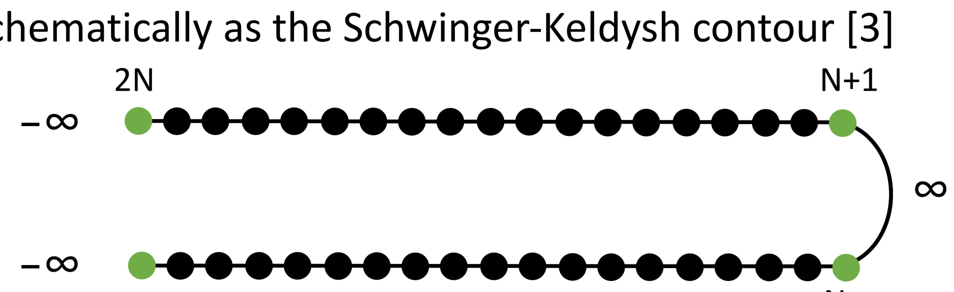


Keldysh Quantum Field Theory

In quantum mechanics systems are described by the density matrix ρ where expectation values are calculated as $\langle O \rangle = \text{Tr}[O\rho]$, while in quantum field theory the key quantity is the partition function $Z = \text{Tr}[\rho]$, where expectation values are given by $\langle O \rangle = \delta Z / \delta \psi_O$. Generically, the density matrix evolves as

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

where U generates time translation. This leads to the presence of two "contours" corresponding to evolving bras and kets respectively. This can be represented schematically as the Schwinger-Keldysh contour [3]



where we have drawn circles to discretize the evolution to finite timesteps and highlighted the boundary times.

For pure states in equilibrium, we can get away with a single contour (since initial and final state are the same)

Summary

The goal is to understand quantum materials by strongly coupling to photon modes and looking at the quantum fluctuations. This can by done using input-output theory where field-theoretical methods give us rigorous perturbation theory results in terms of powers of the non-linear many-body operators that couple arbitrary multiphoton states to a quantum material with strong interactions. This approach can also be used to create effective descriptions of the system's collective modes.

References

[1] Gardiner and Collett Physical Review A 31, 3761 (1985)
[2] Schlawin, Kennes, and Sentef, Applied Physics Rev. 9, 011312 (2022); Dorfman, Schlawin, and Mukamel, Rev. Mod. Physics 88, 045008 (2016)
[3] Kamenev, *Field Theory of Non-Equilibrium Systems*, (2023)
[4] Daniel and Potts, *A Keldysh Path Integral Approach to Input-Output Theory*, Thesis University of Basel (2022)
[5] B. Kass, S. Talkington, A. Srivastava, and M. Claassen, "Many-body photon blockade and quantum light generation from cavity quantum materials" arXiv:2411.08964 (2024)