Non-equilibrium diagrammatic approach
to strongly interacting photons

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Abstract

We develop a non-equilibrium field-theoretical approach based on a systematic diagrammatic expansion for strongly interacting photons in optically dense atomic media. We consider the case where the characteristic photon-propagation range $L_P$ is much larger than the interatomic spacing $a$ and where the density of atomic excitations is low enough to neglect saturation effects. In the highly polarizable medium the photons experience nonlinearities through the interactions they inherit from the atoms. If the atom-atom interaction range $L_E$ is also large compared to $a$, we show that the subclass of diagrams describing scattering processes with momentum transfer between photons is suppressed by a factor $a/L_E$. We are then able to perform a self-consistent resummation of a specific (Hartree-like) diagram subclass and obtain quantitative results in the highly non-perturbative regime of large single-atom cooperativity. Here we find important, conceptually new collective phenomena emerging due to the dissipative nature of the interactions, which even give rise to novel phase transitions. The robustness of these is investigated by inclusion of the leading corrections in $a/L_E$. We consider specific applications to photons propagating under EIT conditions along waveguides near atomic arrays as well as within Rydberg ensembles.
I. INTRODUCTION

The possibility to implement interactions between photons in the quantum regime is recently attracting a lot of interest\cite{1}. One reason is technological, as photon-photon interactions are essential for quantum information processing and would allow to build quantum networks exploiting the ability of photons to efficiently carry information over long distances\cite{2}. Interacting photons are also promising for the creation of synthetic quantum matter, like superfluids\cite{3}, or gapped\cite{4,5} and even topological\cite{6} phases.

From a more fundamental, many-body perspective, an ensemble of strongly interacting photons shows crucial differences from any condensed-matter counterpart and is therefore likely to show novel collective phenomena which have no analog in conventional materials. The first such difference is that the photon number is never conserved so that repumping is needed to compensate losses and reach a driven-dissipative steady state, the latter thus generically being far away from thermal equilibrium. Furthermore, photons do not interact in vacuum and need a material to mediate their mutual interactions. The electromagnetic (EM) modes hybridize with the material giving rise to polaritonic excitations. Here we concentrate on materials made of uncharged but polarizable atoms, where the polaritons (and therefore the photons) inherit their interactions from the latter. This implies a second important feature, namely that the interaction between two photons is a higher order process, requiring the intermediate excitation of the atomic dipoles. Interactions between polaritons in such systems are also naturally long-ranged (as the relevant electromagnetic modes typically extend over many atoms) and retarded (as the time scales of photons and atoms can be respectively tuned to be comparable). Finally, interactions inherited from atomic dipoles can be strongly dissipative due to the spontaneous decay of excited atomic levels. This feature in particular has been shown to be capable of introducing novel many-body phenomena, whereby correlations can be induced by dissipation\cite{9,10,11}.

The implementation of strong interactions between photons in the quantum regime typically requires significant single-photon nonlinearities induced by a large interaction cross-section between a single photon and a single atom\cite{1}, which poses an experimental challenge. It can be overcome by light-confinement via evanescent waves or optical resonators, and/or by providing the atoms with strong, long-ranged interactions preventing multiple atoms to be excited within a large radius, as done by using Rydberg levels.
The theoretical description of such a strongly-interacting, driven-dissipative system of photons in the many-body regime constitutes a challenging task as well. In particular, the large interaction cross sections prevent a perturbative treatment, the driven-dissipative nature does not allow to exploit fluctuation-dissipation relations and prevents for instance the application of Monte Carlo methods, while the long-range interactions additionally hinder an efficient employment of tensor network methods, even in one spatial dimension. A few theoretical approaches have been developed for the few-body regime\textsuperscript{12–16}, while effective field theories have been applied in the many-body regime\textsuperscript{11,17–20}.

Here, we introduce a systematic, diagrammatic approach for the computation of non-equilibrium correlators for a many-body system of strongly interacting photons in an optically dense medium. If the characteristic photon propagation range $L_P$ in the medium is much larger than the spacing $a$ between the atoms, we show that a controlled diagrammatic expansion in powers of $a/L_P$ can be performed, even if the collective light-matter coupling $g_P$ within the mode volume of the photon is large. This perturbative expansion in $a/L_P$ is always valid when the single-atom cooperativity $C_{sa}^p = \left( g_P^2 / \gamma \kappa \right) (a/L_P)$ is much smaller than unity, where $\gamma, \kappa$ are the characteristic dissipation rates of excited atomic levels and photons, respectively. The quantitative validity of our approach can however even be extended to a regime of large single atom cooperativities $C_{sa}^p \gtrsim 1$, provided that the density of atomic excitations is low enough to neglect saturation effects. In such a situation, photons would not experience any nonlinearity or interactions, unless the atoms experience additional, mutual interactions which the photons can inherit. If inter-atomic interactions are present and if their range $L_E/a \gg 1$ is large, we show that the subclass of diagrams describing scattering processes with momentum transfer between photons is suppressed by a factor $\sim a/L_E$ with respect to the remaining Hartree-like diagrams. In this case we are able to perform a self-consistent resummation of the Hartree-like diagram subclass and obtain quantitative results in a strongly non-perturbative regime, which indeed shows important collective behavior and even phase transitions (see also\textsuperscript{21} for a discussion focusing on a specific example).

From a quantum-field-theory perspective, this work constitutes a first attempt to develop a non-relativistic version of Quantum Electrodynamics (QED) where the matter degrees of freedom are dipoles instead of charged electrons, with two further important differences: i) the photons are driven and (partially) confined in space, and ii) the light-matter coupling is far away from the perturbative regime.
In the following, we illustrate specific applications to experiments involving interactions mediated through waveguide photons, for example in photonic-crystal-waveguides\cite{22,23}, as well as Rydberg interactions\cite{21,20}. We consider atomic level structures allowing the photons to propagate under electromagnetically-induced-transparency conditions\cite{30,31}.

The paper is structured as follows: In section II we introduce the \(a/L\)-expansion in general terms, which is then formulated for the specific example of atomic arrays in the vicinity of optical waveguides in section III. After briefly revisiting the phenomenon of electromagnetically induced transparency using our diagrammatic approach (section IV) and the general structure of interactions between polaritons (section V), the implications of strong interactions are discussed, first on the Hartree level (section VI) and finally including all scattering effects to order \(a/L_E\) in section VII. In section VIII we conclude with a short comparison between the case of waveguide-mediated interactions and the case of Rydberg inter-atomic interactions, demonstrating the wide applicability of the presented approach.

II. DIAGRAMMATIC EXPANSION IN THE INVERSE INTERACTION RANGE

The basic idea underlying our diagrammatic approach can be understood in quite general terms. Let us consider a system of two completely different types of particles, which we will for later convenience call photons and atoms. For now we will keep these particles as generic as possible and only fix their mass: Photons are very light (or even massless) and therefore propagate very fast and over long distances, whereas atoms are considered as comparatively heavy, localized and thus slowly moving. Furthermore, neither atoms nor photons shall interact among themselves, that is, excited atoms can only interact via exchange photons and photons only via the non-linear susceptibility of the atomic medium, which results in a Yukawa-type coupling. The stark contrast between the two free theories of atoms and photons allows for a controlled expansion, even in the case of strong collective light-matter interactions. This is due to the large effective mode volume of the photon, suppressing the single-atom cooperativity and thereby providing a useful expansion parameter.

To make this argument more concrete, let us for simplicity consider the specific case of photons with a group velocity \(c\) that couple with a rate \(g\) to the collection of all atoms within an effective mode volume \(L\). We furthermore assume that the atoms are confined to fixed positions in a one-dimensional chain. Furthermore, photons are lost out of the one-
dimensional medium at a rate $\kappa$ and the atomic transition giving rise to a dipole moment takes place between a lower stable level and an excited level decaying at a rate $\gamma$. To avoid the trivial case of the steady state being the vacuum, the atoms are additionally excited by a resonant coherent light source at a Rabi frequency $\Omega$. For this simple model it is easy to write both the atomic and the photonic Green’s functions in a perturbative expansion in the coupling strength $g$. Representing the photonic propagator $G_{\text{ph}}$ with a wavy line and its atomic counterparts $G_{\text{a}}$ with a straight line, one obtains to order $g^2$ the diagrams depicted in Fig. 1.

The leading correction to the bare photonic dispersion due to the atomic medium is given by

\begin{align*}
\text{photons:} & \quad \begin{array}{c}
\begin{array}{c}
\text{atomic transitions}
\end{array}
\end{array} + \begin{array}{c}
\begin{array}{c}
\text{photon-photon correlation}
\end{array}
\end{array}
\end{align*}

\begin{align*}
\text{atoms:} & \quad \begin{array}{c}
\begin{array}{c}
\text{atomic states}
\end{array}
\end{array} + \begin{array}{c}
\begin{array}{c}
\text{atomic interactions}
\end{array}
\end{array} + \begin{array}{c}
\begin{array}{c}
\text{atomic corrections}
\end{array}
\end{array}
\end{align*}

FIG. 1. Illustration of the breakdown of the perturbative expansion of photonic and atomic propagators in second order in the coupling strength $g$. Wavy lines represent photons, straight lines atoms. In the case of a large collective cooperativity the self-energy correction to the photon propagator (second diagram in the first line) becomes large. The same is true for the corrections to the propagator of the atomic state in the second line due to the last contribution, which strongly couples stable and excited atomic states. Large collective cooperativities thus require an improved approach (cfr. Fig. 9).

the polarization bubble shown in Fig. 1a). Performing the corresponding convolutions, one immediately obtains that on-shell each correction (called self-energy in the diagrammatics
jargon) to $G_{ph}$ is proportional to $g^2/\gamma$, which implies that the expansion in powers of $g$ breaks down if the collective cooperativity $C = g^2/(\gamma \kappa)$ becomes of order unity. Equivalently, the last term in Fig. 1b), which is proportional to $\Omega g^2/\gamma \kappa$, becomes larger than the bare coupling $\Omega$ if $C \approx 1$. At the same time however, the other correction to the bare propagator is negligible if the cooperativity per atom $C_{sa} = g^2/(\gamma \kappa L) \ll 1$. Here we have introduced the dimensionless effective mean free path of the photons $L = n c / \kappa$, where $n$ is the number density of the atoms. From the point of view of the atoms this corresponds to the effective interaction range. Thus, if this dimensionless interaction range is large, the small coupling $g/\sqrt{L}$ allows for a partially perturbative treatment, where only a small subclass of diagrams (those that are not suppressed by powers of $1/L$) has to be resummed to all orders, as we illustrate below. With respect to the topology of the diagrams, this expansion is identical to a $1/N$ expansion, where both atoms and photons can appear in $N$ different degrees of freedom (flavors) and the vertex conserves the atomic flavor.

While this expansion is already quite useful, as it allows to introduce controlled interactions between polaritons, it does not immediately allow to enter the regime of strong single atom cooperativities, where the finiteness of the Hilbert space of each atom starts to play an essential role. To correctly account for the finite polarizability of each atom, one additionally has to introduce non-linear Feynman rules, in effect extending the theory to large single atom cooperativities and simultaneously reducing the required set of diagrams to one that – under certain conditions – can be treated exactly (see Sec. [III D]).

### III. APPLICATION GUIDED PHOTONS COUPLED TO ATOMIC ARRAYS

Building on the newly gained understanding that any physical system is suitable for a $1/L$ expansion as long as it exclusively couples degrees of freedom that are well localized in position space to others that are tightly confined in the conjugate momentum space, we will now be more concrete and apply this approach to photons in optical waveguides coupled to an array of atoms. Large single-atom cooperativities in such a setup are for instance reached using atoms trapped within the evanescent-wave of photonic crystal waveguides (PCWs) or tapered-nanofiber waveguide (TNWs). The concepts introduced in this section are however far more general and can be applied in similar ways to any system of interacting polaritons, as will be discussed by a comparison with a gas of Rydberg atoms in
A. The model

We consider a chain of atoms trapped at fixed positions and with the internal level structure shown in Fig. III A. We assume four atomic levels in an $\mathcal{N}$-configuration. The ground state is represented by $|g\rangle$ and the first excited, unstable state by $|e\rangle$. The transition between those levels is (almost) resonant with the energy of a set of propagating photon modes of the waveguide with dispersion $\omega_k^\mathcal{P}$. A further, metastable atomic state $|s\rangle$ can be reached from $|e\rangle$ by stimulated emission of a photon with energy $\omega_L^{(1)}$ into a laser mode, driven at Rabi amplitude $\Omega$. Since photons can be converted into atomic excitations, the EM modes of the waveguide hybridize with the two atomic transitions and give rise to three polariton branches. The $g - e - s$ $\Lambda$-scheme can give rise to electromagnetically-induced-transparency (EIT)\(^{30,31}\), whereby one of the polariton branches becomes almost dissipationless, as described in detail in section IV. The direct photon-photon interaction arising from individual atom saturation is extremely weak\(^{37}\). Such an interaction can be made much stronger by introducing a mechanism for the atoms to interact with one another over a distance. This is achieved via an additional set of exchange-photon modes of the waveguide with dispersion $\omega_k^\mathcal{E}$. These are orthogonally polarized with respect to the $\mathcal{P}$-modes introduced above and can be tuned separately. In particular, it is possible to use the exchange photons to couple a second excited state $|d\rangle$ to the state $|s\rangle$. To adjust the admixture of $|d\rangle$, we introduce a second driving laser of frequency $\omega_L^{(2)}$ and Rabi amplitude $\Omega_s$. In the actual calculations shown here we will for concreteness choose a cosinusoidal dispersion for the $\mathcal{P}$-photons and a quadratic dispersion for the $\mathcal{E}$-photons, which corresponds to the situation in PCWs. The actual choice however does not make any qualitative difference. In general the parabolic approximation to $\omega_k^\mathcal{E}$ is justified by tuning the laser frequency in the vicinity of a dispersion minimum or maximum. In particular, tuning to within the band gap creates a bound state, since the exchange photon cannot propagate and becomes localized around the atom that has emitted it\(^{23}\). This bound state facilitates a strong interaction with other atoms within the region of localization.

Our diagrammatic approach will be formulated within a non-equilibrium functional-
FIG. 2. Level scheme of atoms trapped near a photonic crystal waveguide. External lasers with Rabi amplitudes $\Omega$ and $\Omega_s$ drive transitions between the metastable state $|s\rangle$ and decaying excited states $|e\rangle$ and $|d\rangle$. The two orthogonal polarizations of the photon modes within the PCW are (almost) resonant with the $|g\rangle - |e\rangle$ and $|s\rangle - |d\rangle$ transition respectively. Without the excited state $|d\rangle$ the system therefore reduces to the well known $\Lambda$ level scheme.

integral formalism. However, since for each atom the Hilbert space is finite, more precisely the occupation of all its states sums up to one, the representation of atomic operators in a form that is convenient for the path integral formulation has to be given some thought. Here we will restrict ourselves to the limit of a small density of excited atoms, where saturation effects of the medium can be neglected. As a result, the Schwinger boson representation without explicit restriction to the boson number of each atomic transition will suffice. In particular, the action of the Hamiltonian in the atomic Hilbert space can be broken down into a sum of spin-1/2 operators $\sigma_{\mu,\nu} = |\mu\rangle\langle\nu|$, where $|\mu\rangle, |\nu\rangle \in \{|g\rangle, |e\rangle, |s\rangle, |d\rangle\}$, which can be approximately expressed through bosonic creation and annihilation operators:

$$\sigma_{\mu,\nu} = \hat{a}_\mu^\dagger \hat{a}_\nu .$$

Clearly this approximation allows for an unrestricted occupation of any state of any atom – a shortcoming which will be compensated by the application of non-linear Feynman rules and the restriction to low excitation densities. Since treating spins within a path integral formulation is considerably more complicated than bosons\textsuperscript{38,39}, this transformation is cru-
cial for the tractability of the calculations that lie ahead. Within this linear regime the Hamiltonian part of the system is described by

\[ \hat{H} = \hbar \sum_z \left\{ \omega_e \hat{a}_e^\dagger(z) \hat{a}_e(z) + \omega_s \hat{a}_s^\dagger(z) \hat{a}_s(z) + \omega_d \hat{a}_d^\dagger(z) \hat{a}_d(z) + \left( \Omega e^{-i\omega_L(t)} \hat{a}_e^\dagger(z) \hat{a}_s(z) + h.c. \right) \right. \\
+ \left. \left( \Omega_s e^{-i\omega_L(t)} \hat{a}_d^\dagger(z) \hat{a}_s(z) + h.c. \right) + \int \frac{dk}{2\pi} \left( \omega^P_k [\hat{a}_P(k)]^\dagger \hat{a}_P(k) + \omega^E_k [\hat{a}_E(k)]^\dagger \hat{a}_E(k) \\
+ g_P (\hat{a}_P(k) e^{ikz} u_k^P(z) \hat{a}_e^\dagger(z) \hat{a}_g(z) + h.c.) + g_E (\hat{a}_E(k) e^{ikz} u^E_k(z) \hat{a}_d^\dagger(z) \hat{a}_s(z) + h.c.) \right) \right\}, \]

Where \( u^P_k/E(z) \) represents the periodic/localized part of the Bloch functions of either polarization at quasi-momentum \( k \). Here we use the standard convention for the thermodynamic limit in a crystal with lattice constant \( a = 1 \), namely \( \sum_z e^{ikz} = 2\pi \delta(k) \).

We include the decay of the excited atomic states via

\[ \mathcal{L}_{\gamma_e} \rho = -\hbar \sum_z \frac{\gamma_e}{2} \left\{ \{ \hat{a}_e^\dagger(z) \hat{a}_e(z), \rho \} - 2\hat{a}_e(z)\rho \hat{a}_e^\dagger(z) \right\} \]

\[ \mathcal{L}_{\gamma_d} \rho = -\hbar \sum_z \frac{\gamma_d}{2} \left\{ \{ \hat{a}_d^\dagger(z) \hat{a}_d(z), \rho \} - 2\hat{a}_d(z)\rho \hat{a}_d^\dagger(z) \right\} , \]

which accounts only for independent emission from each atom, neglecting collective effects.\(^{10}\)

We also allow for global photon losses of both polarizations into free space, such as through scattering or absorption

\[ \mathcal{L}_{\kappa_P} \rho = -\hbar \int_k \frac{K_P}{2} \left\{ \{ [\hat{a}_P(k)]^\dagger \hat{a}_P(k), \rho \} - 2\hat{a}_P(k)\rho [\hat{a}_P(k)]^\dagger \right\} \]

\[ \mathcal{L}_{\kappa_E} \rho = -\hbar \int_k \frac{K_E}{2} \left\{ \{ [\hat{a}_E(k)]^\dagger \hat{a}_E(k), \rho \} - 2\hat{a}_E(k)\rho [\hat{a}_E(k)]^\dagger \right\}. \]

Here we have used the notation \( \int_k = L \int \frac{dk}{2\pi} \). Additionally, an incoherent and homogeneous pumping of the propagating modes with a transverse light source shall drive the system out of equilibrium. Without affecting the physics of the dark-state polaritons, one could simply describe this light source by a Markovian bath:

\[ \mathcal{L}_{\kappa_s} \rho = -\hbar \int_k \frac{K_s}{2} \left\{ \left\{ \hat{a}_P(k) [\hat{a}_P(k)]^\dagger + h.c., \rho \right\} - 2 [\hat{a}_P(k)]^\dagger \rho \hat{a}_P(k) - 2\hat{a}_P(k)\rho [\hat{a}_P(k)]^\dagger \right\} \]

The only disadvantage of this description is a large population of non-interacting photons propagating through the system at frequencies far detuned from any atomic resonances. In fact, a transversal light source will not couple to all modes equally well, but due to frequency
dependencies of the mode matching, will predominantly couple to a certain frequency interval. To include this, we instead model the incoherent drive indirectly. First an additional Gaussian mode with a coherence time and length much shorter than the relevant time and length scales for dark-state polaritons is itself driven by a Markovian bath:

\[ \hat{H}_b = \omega_0 \hat{b}^\dagger \hat{b} \quad (7a) \]

\[ \mathcal{L}_{\text{loss}} \rho_b = -\hbar \frac{\kappa_1}{2} \left\{ \{ \hat{b}^\dagger \hat{b}, \rho \} - 2 \hat{b} \rho \hat{b}^\dagger \right\} \quad (7b) \]

\[ \mathcal{L}_{\text{drive}} \rho_b = -\hbar \frac{\kappa_2}{2} \left\{ \{ \hat{b}^\dagger, \rho \} - 2 \hat{b}^\dagger \rho \hat{b} \right\} , \quad (7c) \]

where \( \hat{b}^\dagger \) and \( \hat{b} \) are bosonic creation and annihilation operators for the auxiliary mode with density matrix \( \rho_b \). In a next step this mode then couples bilinearly to the propagating photons:

\[ \hat{H}_{ab} = g_b \int_k (\hat{b}^\dagger \hat{a}_P(k) + \hat{a}_P^\dagger(k) \hat{b}) . \quad (8) \]

In the limit of strong driving, where \( \kappa_1/\kappa_2 \) approaches unity from below, this construction, that effectively mimics a frequency dependent coupling of the system to a highly occupied incoherent bath, is described by the following addition to the Liouvillian

\[ \mathcal{L}_{\kappa_s} \rho(t) = -\frac{\hbar}{2} \int_k \int_{-\infty}^t dt' \left( \kappa_s(t - t') \left[ \hat{a}_{P,I}(k,t)^\dagger, [\hat{a}_{P,I}(k,t'), \rho_I(t')] \right] - \kappa_s(t' - t) \left[ \hat{a}_{P,I}(k,t), [[\hat{a}_{P,I}(k,t'), \rho(t')] \right] \right) , \quad (9) \]

where the additional index \( I \) indicates that operators are to be evaluated in the interaction picture, where \( H \) as well as all Lindblad operators (except \( \mathcal{L}_{\kappa_s} \)) contribute to the time-evolution. Here \( \kappa_s(t) \) has an exponential decay, the parameters of which can be tuned by the properties of the Gaussian mode. While this construction is rather cumbersome if written as Liouvillian, in the path integral description however, the Gaussian mode can be integrated out immediately giving rise to a simple, closed expression for a colored bath.

Note that, in order to control the occupation of the propagating modes, it is sufficient to choose \( \kappa_1 = \kappa_2 \) such that Stokes and anti-Stokes processes are equal in amplitude and then to adjust the loss rate \( \kappa_P \) accordingly. This construction implies that the anti-Stokes processes are at least equally likely as the Stokes processes, such that the bath cannot be inverted and consequently lasing or condensation are excluded\[^{41}\].

The fact that the linearized description of decay of excited atoms violates atom number...
conservation is an unphysical feature of this approximation. Since a more rigorous modeling of spontaneous decay, e.g. via the Lindblad operator $\hat{a}_e \hat{a}^\dagger_e$, is diagrammatically equivalent to a two-body interaction, which significantly complicates a systematic treatment, we compensate these spurious atom losses by fixing the density of atoms in the ground state. As we will see later, as long as saturation effects are negligible, this description of the atoms in combination with a specific selection rule for the Feynman diagrams becomes exact (see Sec. III D).

**B. Non-equilibrium functional-integral formulation on the Keldysh contour**

In order to recast our non-equilibrium problem into a functional-integral form, we choose the real-time Keldysh contour. This contour is directly obtained by writing the expectation value of an operator $\hat{O}$ at a time $t$ by time-evolving the system from the distant past:

$$
\langle \hat{O} \rangle (t) = \frac{\text{Tr} \left( \hat{U}_{t,-\infty} \hat{O} \hat{U}_{-\infty,t} \hat{\rho} (-\infty) \right)}{\text{Tr} (\hat{\rho} (-\infty))}.
$$

(10)

Here $\text{Tr}(\cdot)$ is the trace, $\hat{U}_{t,t'}$ is the time evolution operator from time $t'$ to $t$ and $\hat{\rho} (-\infty)$ is the density matrix of the system in the distant past.

Our goal is to compute the single-particle Green’s functions (GFs) or propagators. Due to our system being driven-dissipative, we cannot assume thermal equilibrium i.e. detailed balance, such that there are in principle two independent propagators, the retarded

$$
iG^R_{ij}(x,x') = \theta(t-t') \left\langle \left[ \hat{a}_i(x), \hat{a}^\dagger_j(x') \right] \right\rangle,
$$

(11)

and the Keldysh GF

$$
iG^K_{ij}(x,x') = \left\langle \left\{ \hat{a}_i(x), \hat{a}^\dagger_j(x') \right\} \right\rangle,
$$

(12)

with $i,j = g,e,s,d,E,P$ labeling the degree of freedom and $x = (z,t)$ being the space-time coordinate. We now treat the time-evolution of these expectation values by means of the coherent state path integral. In doing so one inserts resolutions of unity in terms of coherent states spaced in infinitesimal timesteps along the time-evolution. Evaluation of the resulting matrix elements then replaces the operators $\hat{a}(x)_j$ and $\hat{a}^\dagger(x)'_j$ by the field $a(x)_j$ and its complex conjugate $\bar{a}(x)_j$. However according to (10), one has to evolve the system both forward and backward in time, which requires us to split each field into a part on the
forward branch (denoted with a superscript +) and one on the backward branch (labeled by a −), whereby the GFs are now given by

\[ iG^R_{ij}(x, x') = \theta(t - t') \left( \langle a^+_i(x) \bar{a}^+_j(x') \rangle - \langle a^+_i(x) \bar{a}^-_j(x') \rangle \right) \]
\[ iG^K_{ij}(x, x') = \langle a^+_i(x) \bar{a}^+_j(x') \rangle + \langle a^+_i(x) \bar{a}^-_j(x') \rangle . \]

Once one performs the so called Keldysh rotation to quantum and classical fields

\[ a^q_j(x) = \frac{1}{\sqrt{2}} (a^+_j(x) - a^-_j(x)) \]
\[ a^{cl}_j(x) = \frac{1}{\sqrt{2}} (a^+_j(x) + a^-_j(x)) , \]

of which the former can have identically vanishing correlations: \( \langle a^q_i(x) \bar{a}^q_j(x') \rangle \equiv 0 \), the Keldysh and retarded GFs take the much simpler forms

\[ iG^R_{ij}(x, x') = \langle a^{cl}_i(x) \bar{a}^q_j(x') \rangle \]
\[ iG^K_{ij}(x, x') = \langle a^{cl}_i(x) \bar{a}^{cl}_j(x') \rangle . \]

Since additionally the advanced GF \( G^A(x, x') = \langle a^q_i(x) \bar{a}^{cl}_j(x') \rangle \) satisfies \( G^A_{ij}(x, x') = [G^R_{ij}]^*(x', x) \), where \((\cdot)^*\) denotes the complex conjugation, no further independent propagators exist. For the non-interacting atoms coupled to the coherent laser fields, the inverse retarded GF reads

\[
\left[ \tilde{G}^R_{a,0} \right]^{-1} (\omega, \omega') = \begin{pmatrix}
(\omega - \omega_d + i \frac{\gamma_d}{2}) \delta(\omega - \omega') & -\Omega_s \delta(\omega - \omega' - \omega^{(2)}_L) & 0 & 0 \\
-\Omega_s \delta(\omega - \omega' + \omega^{(2)}_L) & (\omega - \omega_s + i \frac{i}{2}) \delta(\omega - \omega') & -\Omega \delta(\omega - \omega' + \omega^{(1)}_L) & 0 \\
0 & -\Omega \delta(\omega - \omega' - \omega^{(1)}_L) & (\omega - \omega_e + i \frac{i}{2}) \delta(\omega - \omega') & 0 \\
0 & 0 & 0 & (\omega + i \frac{i}{2}) \delta(\omega - \omega')
\end{pmatrix},
\]

where we used the basis

\[ a^{(q,cl)}_a(\omega, z) = \begin{pmatrix}
a_d(\omega, z) \\
a_s(\omega, z) \\
a_e(\omega, z) \\
a_g(\omega, z)
\end{pmatrix}^{(q,cl)} . \]

However as it turns out, it is far more convenient to transform into a rotating frame, where the states \(|e\rangle, |s\rangle\) and \(|d\rangle\) rotate at frequencies \(\omega_e, \omega_e - \omega^{(1)}_L\) and \(\omega_e - \omega^{(1)}_L + \omega^{(2)}_L\) respectively.
Within this frame the atomic GF becomes time translationally invariant, that is \( G_{a,0}^{-1}(\omega, \omega') = G_{a,0}^{-1}(\omega) \delta(\omega - \omega') \) with

\[
[G_{a,0}^{R}]^{-1}(\omega) = \begin{pmatrix}
\omega - \Delta_d - \Delta_s + i \frac{\gamma_d}{2} & -\Omega_s & 0 & 0 \\
-\Omega_s & \omega - \Delta_s + i \frac{\gamma_e}{2} - \Omega & 0 \\
0 & -\Omega & \omega + i \frac{\gamma_e}{2} & 0 \\
0 & 0 & 0 & \omega + i \frac{\gamma_e}{2}
\end{pmatrix},
\]

and the fields shifted accordingly in frequency:

\[
a^{(q,cl)}_a(\omega, z) = \begin{pmatrix}
a_d(\omega + \omega_e - \omega_L^{(1)} + \omega_L^{(2)}, z) \\
& a_s(\omega + \omega_e - \omega_L^{(1)}, z) \\
& a_e(\omega + \omega_e, z) \\
& a_g(\omega, z)
\end{pmatrix}^{(q,cl)}.
\]

Here the detunings \( \Delta_s = \omega_e - \omega_L^{(1)} - \omega_s \) and \( \Delta_d = \omega_d - \omega_s - \omega_L^{(2)} \) between laser frequencies and atomic transitions have been introduced. In order to avoid confusion, throughout the remainder of this manuscript we will exclusively work in the rotating frame. The corresponding Keldysh component of the inverse GF within the same frame of reference is then given by

\[
D^K_{a,0}(\omega) = \begin{pmatrix}
 i\gamma_d & 0 & 0 & 0 \\
 0 & i\epsilon & 0 & 0 \\
 0 & 0 & i\gamma_e & 0 \\
 0 & 0 & 0 & (3 - 2n_V)i\epsilon
\end{pmatrix}.
\]

It should be pointed out, that the factor \( 3 - 2n_V \) in the ground-state sector accounts for the occupation of this mode with a homogeneous number density of lattice defects or vacancies \( n_V \in [0,1] \). Thus for \( n_V = 0 \) the ground-state is homogeneously occupied with one atom per site \( (n = -1/2 + i \int \frac{d\omega}{4\pi} G^K = 1 - n_V = 1) \).

An equivalent rotation can also be performed for the retarded and Keldysh component of the inverse photon GF, which then are given by

\[
[G_{p,0}^{R}]^{-1}(\omega, k) = \begin{pmatrix}
\omega - \Delta_E(k) + i \frac{\kappa_E}{2} & 0 \\
0 & \omega - \Delta_P(k) + i \frac{\kappa_P}{2}
\end{pmatrix}.
\]
and
\[ D_{p,0}^{K}(\omega, k) = \begin{pmatrix} i\kappa_{E} & 0 \\ 0 & i\kappa_{P} + 2i\kappa_{s}(\omega) \end{pmatrix} \] (23)
respectively. Here we expressed both functions in the basis
\[ a_{p}^{(q,cl)}(\omega, k) = \begin{pmatrix} a_{E}(\omega + \omega_{L}^{(2)}, k) \\ a_{P}(\omega + \omega_{e}, k) \end{pmatrix}^{(q,cl)}, \]
(24)
and introduced the detunings \( \Delta_{P}(k) = \omega_{P}(k) - \omega_{e} \) and \( \Delta_{E}(k) = \omega_{E}(k) - \omega_{L}^{(2)} \). Note that here we have already performed the Gaussian integration over the auxiliary field \( b \), after which the inverse probe photon propagator in general is modified by the subtraction of \( g_{b}^{2}G_{b}(\omega) \).

Assuming very strong coupling to the incoherent source, however, \( \kappa_{1} \) and \( \kappa_{2} \) diverge, while \( \kappa_{0} = 2(\kappa_{1} - \kappa_{2}) \) and \( \kappa_{s} = 2g_{b}^{2}\kappa_{1} \) are kept finite. In this limit \( g_{b}^{2}G_{b}^{R}(\omega) \) vanishes, while \( \kappa_{s}(\omega) = -ig_{b}^{2}G_{b}^{K}(\omega)/2 = \kappa_{s}/((\omega - \omega_{0})^{2} + \kappa_{0}^{2}) \) remains finite.

Modeling the situation in PCWs, throughout this work we will approximate the dispersion of the exchange photons as parabolic: \( \omega_{E}(k) = \omega_{E}^{0} - \alpha_{E}(k - k_{E})^{2} \) around the band edge \( \omega_{E}(k_{E}) = \omega_{E}^{0} \), which is assumed to be slightly detuned against the \( |s\rangle - |d\rangle \) transition. With this choice the exchange photon spreads diffusively around the emitting atom with an average mode volume and therefore interaction range given by
\[ L_{E} = \sqrt{\alpha_{E}/\kappa_{E}}. \] (25)

On the other hand, since we are eventually interested in the interaction induced modifications to the dispersion of the propagating photons, we will require no approximations to the dispersion \( \omega_{k}^{P} \). As already stressed above, the actual form of the photon dispersion does not play a qualitative role.

Making use of the above notation, the non-interacting part of the action \( S = S_{0} + S_{\text{int}} \) can be fully expressed in terms of the bare atomic (subscript \( a \)) and photonic (subscript \( p \)) GFs as
\[ S_{0} = \hbar \int \frac{d\omega}{2\pi} \left( \sum_{z} [a_{a}]^{*}(\omega, z)G_{a,0}^{-1}(\omega)a_{a}(\omega, z) + \int \frac{dk}{2\pi} [a_{p}]^{*}(\omega, k)G_{p,0}^{-1}(\omega, k)a_{p}(\omega, k) \right). \] (26)

Here the index 0 is meant to indicate the absence of self-energy corrections due to interactions (see below). Furthermore \( a_{\mu} = \{a_{\mu}^{a}, a_{\mu}^{p}\} \) with \( \mu \in \{a, p\} \) are the vectors of classical and
quantum fields with the corresponding inverse Keldysh matrix GFs given by
\[
G_{\mu,0}^{-1} = \begin{pmatrix} 0 & \left[ G^A_{\mu,0} \right]^{-1} \\ \left[ G^R_{\mu,0} \right]^{-1} & D^K_{\mu,0} \end{pmatrix}.
\] (27)

Finally, the interaction part of the action reads
\[
S_{\text{int}} = \int \frac{d\omega}{2\pi} \int \frac{dk}{2\pi} \sum_z \left( \frac{1}{\sqrt{2}} g_{PE} e^{ikz} u^P_k(z) \left[ a^\alpha_k(k) \left( \bar{a}^\alpha_{g}(z)a^a_{g}(z) + \bar{a}^c_{g}(z)a^cl_{g}(z) \right) + a^c_{g}(z) \left( \bar{a}^\alpha_{g}(z)a^a_{g}(z) - \bar{a}^c_{g}(z)a^cl_{g}(z) \right) \right] + \text{h.c.} + \frac{1}{\sqrt{2}} g_{BE} e^{ikz} u^E_k(z) \left[ a^E_{k}(z) \left( \bar{a}^\alpha_{d}(z)a^a_{s}(z) + \bar{a}^c_{d}(z)a^cl_{s}(z) \right) + a^c_{d}(z) \left( \bar{a}^\alpha_{d}(z)a^a_{s}(z) - \bar{a}^c_{d}(z)a^cl_{s}(z) \right) \right] + \text{h.c.} \right). \] (28)

As the atoms are fixed at positions commensurate with the PCW, we can use the periodicity of the dimensionless Bloch functions \( u^{P,E}_k(z) \) to replace them by \( u^{P,E}_k(0) \). In general, careful engineering of the PCW allows some control over the momentum dependence of \( u^{P,E}_k(0) \). Here we will choose the simplest approximation of a constant, which we then absorb into the coupling via the replacement \( g_{P,E} u^{P,E}_k(0) \to g_{P,E} \).

As in equilibrium theory, one can apply Wick's theorem to find the dressed GFs
\[
G^{\alpha\beta}_{\mu}(x, x') = \langle \left[ a^\alpha_{\mu}(x) \ast \bar{a}^\beta_{\mu}(x') \right] \rangle_S, \] (29)

with \( \ast \) the outer product and \( \mu \in \{a, p\} \) as well as \( \alpha, \beta = \text{cl, q} \). Here, as opposed to the bare propagators, the expectation value is taken with respect to the full action \( S \). Expanding the exponent \( e^{iS_{\text{int}}} \) under the path integral, one obtains the infinite Dyson series
\[
G = G_{\mu,0} + G_{\mu,0} \circ \Sigma_{\mu} \circ G_{\mu,0} + G_{\mu,0} \circ \Sigma_{\mu} \circ G_{\mu,0} \circ \Sigma_{\mu} \circ G_{\mu,0} + \ldots, \] (30)

where \( \circ \) denotes the convolution in space and time with a simultaneous matrix product in the Keldysh index \( \{\text{cl, q}\} \) as well as the field components \( g, e, s, d \) or \( P, E \) for the atomic \( (\mu = a) \) or photonic propagators \( (\mu = p) \) respectively. Summation of this geometric series for the retarded GF gives the same result as in equilibrium theory
\[
G^R_{\mu} = \left( G^R_{\mu,0} - \Sigma^{R}_{\mu} \right)^{-1}. \] (31)

For the Keldysh component however one finds
\[
G^K_{\mu} = G^R_{\mu} \circ \left( \Sigma^{K}_{\mu} - D^K_{\mu,0} \right) \circ G^A_{\mu}. \] (32)

As \( \Sigma^{R,K}_{\mu} \) in general depend on Keldysh and retarded components, these two Dyson equations are coupled and have to be solved simultaneously.
C. Kramers-Kronig relations

Due to causality, each vertex involves either one or three quantum fields. There are thus four copies of each vertex, differing only in the Keldysh index while otherwise being identical. As every retarded GF has to connect a quantum and a classical field, while the Keldysh component connects only classical fields, the distinction in the number of quantum indices in a vertex results in it being connected to different components of the matrix GF. In general, this gives rise to a large amount of Feynman diagrams to be calculated. Luckily, most of these can be related to one another using Kramers-Kronig relations. In the following we will illustrate how these relations can be exploited. For the sake of compactness of the graphical illustrations, we avoid drawing all possible diagrams so that each line we draw can be a retarded, advanced or Keldysh GF, as long as the vertex allows it.

It is useful to introduce the distribution function $F(\omega)$, which encodes how strongly the effective environment tries to populate a certain degree of freedom. It is defined by $G^K(\omega) = G^R(\omega)F(\omega) - F(\omega)G^A(\omega) = 2i\Im(G^R(\omega))F(\omega)$, where the last equality holds only in a scalar theory, or if all GFs can be diagonalized simultaneously. In the absence of particles $F(\omega) = 1$ allows to express the Keldysh component through the retarded GF. We will denote this special case of an unoccupied degree of freedom by a subscript 0 attached to the causality index ($K$) as in $G^{K_0}$. For these empty modes, invoking Kramers-Kronig relations for the retarded GF

\begin{align}
\Re G^R(\omega) &= \mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega'}{\pi} \frac{\Im G^R(\omega')}{\omega' - \omega} \\
\Im G^R(\omega) &= -\mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega'}{\pi} \frac{\Re G^R(\omega')}{\omega' - \omega}
\end{align}

(33)

allows to find some simplifications. Indeed, any two GFs $G_1(\omega)$ and $G_2(\omega)$ will obey

\begin{align}
\int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} G^{K_0}_1(\omega')G_2^R(\omega - \omega') &= \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} G^R_1(\omega')G^{K_0}_2(\omega - \omega') \\
\int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} G^{K_0}_1(\omega')G_2^R(\omega + \omega') &= -\int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} G^A_1(\omega')G^{K_0}_2(\omega + \omega'), \quad (34)
\end{align}

which follow immediately from (33) by splitting the retarded GF into real and imaginary
part. Additionally, one has
\[
\int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} G_1^{K}(\omega') G_2^{K}(\omega - \omega') = \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \left( G_1^{A}(\omega') G_2^{A}(\omega - \omega') + G_1^{R}(\omega') G_2^{R}(\omega - \omega') \right)
\]
\[
\int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} G_1^{K}(\omega') G_2^{K}(\omega + \omega') = -\int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \left( G_1^{A}(\omega') G_2^{R}(\omega + \omega') + G_1^{A}(\omega') G_2^{R}(\omega + \omega') \right),
\]
which are easily proven by applying the Fourier transform to the convolution and using that

\[
\mathcal{F}(\mathcal{H}(f)) = -i \text{sgn}(t) \mathcal{F}(f),
\]

where \( f \) is any function and \( \mathcal{H} \) and \( \mathcal{F} \) are Hilbert and Fourier transform, respectively.

Similar identities, using the same method, can also be proven for more complicated products and higher order convolutions of GFs. Since the bare atomic propagators for all but the ground state, as well as that of the bare exchange photon, are unoccupied, we can use these identities to great effect in the following calculations.

### D. Non-linear Feynman rules

Apart from the Keldysh structure, the interaction part of the action given in (28) contains two different types of vertices, both depicted in Fig. 3, which under time reversal pairwise transform into one another. Either an atom is excited through the absorption of a photon (see Fig. 3a), or, by the time-reversed process of emitting a photon, the atom returns to a stable state (c.f. Fig. 3b). Using the vertices, one can draw up all Feynman diagrams order by order in the coupling constants. However, in doing so one applies bosonic Feynman rules to atoms, which due to the strong nonlinearities should instead have a restricted Hilbert space with \( \sum_{j=g,e,s,d} \langle \hat{a}_j^\dagger \hat{a}_j \rangle = 1 \). This implies that each atom occupies either only one level or in general a properly restricted superposition. One therefore has to be careful not to over-count diagrams by simultaneously placing an atom in the same state twice (which would be allowed for bosons). This means that, at every point in time and in every diagram, two counter propagating atomic lines belonging to the same atom have to be found in distinct levels, or must otherwise be identified with one another, i.e. their lines in the Feynman diagram have to be contracted.

In general it is very hard to fully enforce these conditions, as one would need to implement increasingly complicated restrictions in real-time on each and every perturbation to the bare
scalar GFs. Doing so for all diagrams would eventually restore the exact, finite Fock space of the atoms. Here, we instead limit ourselves to impose restrictions allowing to exactly compute the fully dressed, single probe photon propagator in the absence of the state $|d\rangle$.

As we will see, the insertion of self-energies in the form of polarization bubbles – which are diagrams of the type shown on the right of Fig. 1a) – into the bare probe photon GF will hybridize this propagating photon mode with stationary atoms, forming polaritons in the process. Without state $|d\rangle$, and without saturation effects these polaritons will not interact among each other. When introducing polariton-polariton interactions via coupling to $|d\rangle$ it is then of paramount importance to expand around the correct limit of non-interacting polaritons, which will only be ensured by the implementation of the above restrictions imposed by non-linear Feynman rules.

In the non-interacting regime, where the polariton self-energy is given by a polarization bubble with the external laser fields mixing states $|e\rangle$ and $|s\rangle$ and the probe photons mixing $|g\rangle$ and $|e\rangle$, it suffices to demand that any two counter-propagating GFs of the same atom have to involve disjoint sets of states. All diagrams where this is not the case are simply set to zero.

We now show that these simplified non-linear selection rules correctly capture the retarded polariton GF. The latter reads

$$G_{\text{RP}}^R(\omega, k) = \left( G_{\text{RP}}^R(\omega, k)^{-1} - \Sigma_{\text{RP}}^R(\omega) \right)^{-1}, \quad (37)$$

with the self-energy given by

$$\Sigma_{\text{RP}}^R(\omega) = \frac{ig^2_P}{2} \int \frac{d\omega'}{2\pi} \left( G^K_{e}(\omega + \omega')G^A_g(\omega') + G^K_{e}(\omega + \omega')G^K_g(\omega') \right). \quad (38)$$

We now make use of the Kramers-Kronig relations [34] and realize that only diagrams with either $F_g(\omega) \neq 1$ or $F_e(\omega) \neq 1$ are finite, and thus

$$\Sigma_{\text{RP}}^R(\omega) = \frac{ig^2_P}{2} \int_{\infty}^{\infty} \frac{d\omega'}{2\pi} \left( \delta G^K_{g}(\omega')G^R_{e}(\omega + \omega') + G^A_g(\omega')\delta G^K_{e}(\omega + \omega') \right), \quad (39)$$

where $\delta G^K(\omega) = G^K(\omega) - 2i\Delta G^K(\omega)$ is related to the spectral number density by $n(\omega) = i\delta G^K/2$. However, as the atomic medium without probe photons is entirely in the ground state and no atoms are being created, the only way to get $\delta G^K_{e}(\omega) \neq 0$ is by coupling to $\delta G^K_{g}(\omega)$. On the other hand, corrections to the bare ground-state propagator all inevitably have to involve the excited state $|e\rangle$. 

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To compare the effect of the exact and simplified non-linear Feynman rules, consider the perturbative insertion of corrections into the bare retarded GFs:

$$G^R(t,t') = G_0^R(t,t') + \int dt_1 \int dt_2 G_0^R(t,t_1) \Sigma^R(t_1,t_2) G_0^R(t_2,t') + \ldots , \quad (40)$$

which due to causality are non-zero only if $t > t_1 > t_2 > t'$. Consequently, none of the GFs and self-energies can be evaluated simultaneously and no cancellations due to the non-linear Feynman rules are required. Similarly, the Keldysh component of the interacting GF is given by

$$\delta G^K(t,t') = \int dt_1 \int dt_2 G^R(t,t_1) (\delta \Sigma^K(t_1,t_2) - \delta D^K_0(t_1,t_2)) G^A(t_2,t'), \quad (41)$$

where $\delta D^K_0 = D^K_0 - 2i \Im \left( [G_0^R]^{-1} \right)$ and $\delta \Sigma^K(t_1,t_2) = \Sigma^K(t_1,t_2) - 2i \Im \Sigma^R(t_1,t_2)$ have been introduced. Due to the retarded and advanced GFs one has $t > t_1$ and $t' > t_2$. Clearly those insertions with $t_1 < t'$ have to be discarded, as then, between these times, the retarded and advanced GF of the same state counter-propagate. With this restriction in place $\delta \Sigma^K(t_1,t_2)$ has to be evaluated at $t'$, which is necessarily simultaneous with the retarded GF of the other state in the polarization bubble, and the diagram again has to be removed. In the end, as only the ground-state satisfies $\delta D^K_0 \neq 0$, we are left with the simple result

$$\Sigma^K_R(\omega) = \frac{ig^2}{2} \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \delta G^K_{g,0}(\omega') G^R_{e}(\omega + \omega') , \quad (42)$$

where in $\Sigma^K_R$ no dependence on $G_g$ is allowed. For the Keldysh component of the polariton self-energy one has, due to the Kramers-Kronig relations $^{35}$

$$\Sigma^K_P(\omega) = \frac{ig^2}{2} \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \delta G^K_{g}(\omega') \delta G^K_{e}(\omega + \omega') . \quad (43)$$

Following a similar argument as above, one can show that this contribution vanishes once either the full or the simplified non-linear selection rule is applied. As these arguments can be continued order by order in the coupling constants, we find that for non-interacting polaritons both selection rules coincide. For an alternative proof that electromagnetically induced transparency in the limit of low polariton densities is exactly recovered by the simplified non-linear Feynman rules see Appendix XI A.

For higher order diagrams of the probe photon propagator that involve the exchange photon, as well as for the polarization bubble of the exchange photon itself, these new simplified
Feynman rules do not work quite as well. The reason for this lies in the fact that both states $|s\rangle$ and $|d\rangle$ that necessarily appear in the polarization bubble of an exchange photon, have non-vanishing self-energies. In real time these insertions into the bare propagators can then partially exclude each other, meaning that the simplified non-linear Feynman rules no longer correctly capture the polarizability of the atoms. However, if the effective coupling rate between states $|s\rangle$ and $|d\rangle$ is small compared to $\gamma_d$, the excited atom will likely have decayed before it can be transferred into another state. To ensure this, we will exclusively work in a regime of small $\Omega_s/\gamma_d$. Note however, that this condition will be significantly modified upon inclusion of strong interpolariton interactions, wherefore we will also require $(\Omega_s^{\text{eff}})^2/(\gamma_d^{\text{eff}} \gamma_s^{\text{eff}}) \ll 1$ for the fully dressed quantities.

In order to test that the choice of the specific implementation of the non-linear Feynman rules – of which many different versions are available – does not affect the results, we compare the two extreme options. One is the most strict implementation of the Feynman rules, where all diagrams that could at least partially be forbidden are entirely excluded. The other option corresponds to the opposite choice, where all at least partially allowed diagrams are fully included. In the following, we will refer to these two options as the “strict” and “lenient” implementation of the Feynman rules. If we observe no difference between the results from both options, the ambiguity in the non-linear Feynman rules is of no quantitative significance and either version can be used to provide a lowest order approximation to the actual (time-dependent) selection rules.

In summary, the nonlinear Feynman rules outlined here partially compensate the unphysical tendency of the bosonized atoms to bunch together with the photons. As long as the number density of excited atoms is small compared to that of the ground state, saturation effects of the atomic medium can be neglected and no further selection rules have to be implemented. While this restriction to the selection of diagrams might seem complicated to

\[
\begin{align*}
\text{a)} & \quad \begin{tikzpicture}[baseline={([yshift=-.5ex]current bounding box.center)}]
    \node (f1) at (0,0) {$|g\rangle$};
    \node (f2) at (1,0) {$|c\rangle$};
    \node (f3) at (2,0) {$|s\rangle$};
    \node (f4) at (3,0) {$|d\rangle$};
    \node (f5) at (4,0) {$|\rangle$};
    \draw[->] (f1) -- (f2);
    \draw[->] (f2) -- (f3);
    \draw[->] (f3) -- (f4);
    \draw[->] (f4) -- (f5);
    \node at (2,1) {$\omega_k^P$};
    \node at (2,-1) {$\omega_k^G$};
\end{tikzpicture} \\
\text{b)} & \quad \begin{tikzpicture}[baseline={([yshift=-.5ex]current bounding box.center)}]
    \node (f1) at (0,0) {$|c\rangle$};
    \node (f2) at (1,0) {$|g\rangle$};
    \node (f3) at (2,0) {$|s\rangle$};
    \node (f4) at (3,0) {$|d\rangle$};
    \node (f5) at (4,0) {$|\rangle$};
    \draw[->] (f1) -- (f2);
    \draw[->] (f2) -- (f3);
    \draw[->] (f3) -- (f4);
    \draw[->] (f4) -- (f5);
    \node at (2,1) {$\omega_k^P$};
    \node at (2,-1) {$\omega_k^G$};
\end{tikzpicture}
\end{align*}
\]

FIG. 3. Yukawa-type interaction vertices form the fundamental building blocks of QED with neutral atoms and guided photons.
enforce consistently, we will see that it actually simplifies the Feynman diagrams.

E. Loop reduction

Beyond the Kramers-Kronig relations, a further significant simplification can be achieved by noting that the atomic ground-state has no dynamics of its own. Hence any loop involving the bare Keldysh component of the ground-state propagator can be computed trivially\textsuperscript{45}. In and of itself this is not a particularly useful observation. In combination with the Kramers-Kronig relations and the non-linear Feynman rules introduced in the last subsections, however, several loop integrals can be computed exactly.

To better understand how all of these properties come together, let us consider the case of a probe photon propagating through any polarizable medium. This process to leading order in \(1/L_P\) is described by the diagrammatic equation in Fig.\textsuperscript{4} Here the propagator of the excited state cannot be specified further, since interactions with other excited atoms can and will dress it. The ground-state propagator on the other hand only couples to other states via the absorption of a probe photon. Employing the Feynman rules of section III D, it will thus always be described by the bare GF. Consequently, the diagram of Fig.\textsuperscript{4} for the retarded probe photon propagator is solved by \textsuperscript{37} and \textsuperscript{42} Since furthermore \(G_{g,0}^K(\omega) = -2\pi i(3 - 2n_V)\delta(\omega)\), the remaining integral can then be solved immediately, such that one finally obtains

\[
\Sigma_P^R(\omega) = g_P^2(1 - n_V)G_e^R(\omega),
\]  

(44)
completely independent of the form of the interactions between excited states. A similar
calculation gives the equivalent result
\[ \Sigma^K_P(\omega) = g_P^2 (1 - n_V) \delta G^K_e(\omega) \] (45)
for the Keldysh component of the self-energy. Hence, the result for the polarization bubble
is the same as for a bilinear coupling converting a photon into an excited atomic state, albeit
with the modified coupling constant \( \tilde{g}_P = g_P \sqrt{1 - n_V} \).
This identification changes the topology of diagrams. However, quite importantly the or-
dering in powers of the inverse interaction range remains unaffected.
Note that, despite the extremely long life time of the meta-stable state \( |s\rangle \), due to the static
laser field coupling to states \( |e\rangle \) and \( |d\rangle \) the corresponding Keldysh component \( G^K_s(t - t') \)
explicitly depends on time and a similar identity for particle hole loops involving \( |s\rangle \) is not
quite as useful.

F. Self-consistence and conserving approximations

In studying out-of-equilibrium interacting problems within a diagrammatic approach the
self-consistent solution of the Dyson equations becomes crucial for two main reasons. Firstly,
the long-time behavior and in particular the steady state cannot be described perturbatively.
Secondly, the integrals of motion of the problem are only correctly included within the so
called conserving approximations, which themselves can be derived from an appropriate
thermodynamic functional and always result in self-consistent theories, where all self-energy
insertions are repeated in every internal GF.
In the present case it is unfortunately impossible to build a proper functional, since it
would be irreconcilable with the approximate non-linear Feynman rules introduced above.
Having a conserving approximation in our case is however not crucial. This is a consequence
of the incoherent, transversal drive and Markovian losses. These neither conserve energy
nor quasi-momentum. Therefore, the only conserved quantity is the atom number, which
we approximately enforce, at least on average, by means of the non-linear Feynman rules.
While dropping these would allow to construct a self-consistent functional, the resulting
theory would consequently not conserve the atom number either. We will largely make
use of self-consistent solutions of our Dyson equations in order to include the important
non-perturbative effects.

**IV. LEADING ORDER: POLARITONS AND ELECTROMAGNETICALLY INDUCED TRANSPARENCY**

As was already explained in Sec. III E if there is no interaction between different atoms, other than via the exchange of probe photons, the probe photon propagator is fully given by the solution to the diagrammatic equation in Fig. 4. Of course this is only true in the case of a low excitation density in the atomic medium, since otherwise saturation effects will induce further interactions between probe photons that are not captured by the present approach (see Appendix XI A). In this low excitation density limit however, the retarded photon propagator $G_P$ for $g_E = \Omega_s = 0$ can be directly obtained from Eqs. 37 and 44, where

$$G_e^R(\omega) = G_{e0}^R(\omega) = 1 \over \omega - \Omega^2/\omega - \Delta_e + i\gamma_e/2 + i\epsilon/2$$

$$G_e^K(\omega) = G_{e0}^K(\omega) = -2i\Im G_{e0}^R(\omega)$$

are the components of the bare propagators of the excited state $|e\rangle$ obtained by inverting $[G_{a0}^R]^{-1}$ in Eq. 19. Without any coupling to state $|d\rangle$ the atomic level scheme is identical to the well known Λ-scheme, which in the limit of vanishing excitation density has been solved exactly $^{30}$ and can exhibit electromagnetically induced transparency (EIT). One should note that the solution we give here involves no approximations beyond the linearization of the spin degree of freedom, which we showed in Sec. III D to be fully compensated by simple non-linear Feynman rules. As such, it is not surprising that the result will be identical with that obtained in $^{30}$.

As the physics of EIT will be important for the phenomenology presented in the following, we will summarize its main features in the rest of this section despite them being well known. This also gives us the opportunity to showcase our formalism in a familiar setting. One can immediately identify $G_e^R$ with the polarizability of the medium. Hence, as hinted to earlier, $G_P^R$ no longer describes free photons, but the eigenmodes of the system, which are photons hybridized with the medium. The dispersion of these new degrees of freedom, the so-called polaritons, has three branches resulting from the coupling of two atomic transitions and the photonic dispersive mode, which far away from the atomic resonance $\Delta_e$ is essentially that of the free photon. Due to the vanishing losses of state $|s\rangle$, however, the central branch –
the so-called dark-state polariton, which is a combination of a photon and an atom in state $|s\rangle$ without any admixture of the lossy $|e\rangle$ – is very long lived. The absence of an admixture of excited state in the dark-state polariton can be explained by a destructive interference between two pathways. Since the direct excitation $|g\rangle \rightarrow |e\rangle$ has the same probability (but opposite phase) as the indirect process $|g\rangle \rightarrow |e\rangle \rightarrow |s\rangle \rightarrow |e\rangle$, no population is transferred to $|e\rangle$. This interpretation becomes apparent by inspecting the diagrammatic expression for $G_e$ shown in Fig. 5.

Since the dark-state polariton is a linear superposition of a localized atom and propagating photon, its group velocity can be tuned between 0 and that of the bare photon by adjusting the ratio $\Omega/g_P$. For $\Omega \rightarrow 0$ the polariton stops, however as the polariton slows down losses are reduced at the same rate, such that the penetration depth of photons into the waveguide is not affected. This can be easily verified by comparing the group velocity of the dark-state polariton with its line-width. Linearizing the dispersion of the free photons, which on the energy scale of the susceptibility of the medium (set by $\gamma_e$) is typically well justified, the group velocity can be determined from the pole of the polariton GF $G_P^R(\omega, k)$ given by Eqs. (37)(44) and (46). In the limit of mostly atomic polaritons, where the ratio between atomic and photonic contributions to the polariton $\theta = g_P^2(1 - n_V)/\Omega^2$ becomes large, an expansion around the EIT window results in the condition

$$
\left[ G_P^R(\omega, k) \right]^{-1} = \frac{1}{\theta} (\omega - \Delta_s) - v_P(k - k_{EIT}) + i\eta(\omega - \Delta_s)^2 + i\kappa_P/2 = 0 ,
$$

where $v_P$ is the local group velocity of the bare photon near the resonance at $k = k_{EIT}$ with the laser acting on the $|s\rangle - |e\rangle$ transition. Furthermore we have introduced the convenient abbreviation $\eta = \gamma_e\theta/(2\Omega^2)$. At the center of the EIT window the group velocity is given by

$$
v_g = \frac{d\omega_{res}}{dk} = \frac{v_P}{\sqrt{\theta^2 + 2\eta\kappa_P}} \sim \Omega^2 ,
$$

![FIG. 5. Dyson equation for $G_e$ in the polarization bubble. The interference between the direct excitation of an atom to $|e\rangle$ and the indirect path via $|s\rangle$ gives rise to EIT.](image)
where $\omega_{\text{res}}$ satisfies the condition \(^47\). On the other hand, at $k_{\text{EIT}}$ the line-width of the dark-state polariton is given by

$$\Delta\omega = \frac{\sqrt{-\theta^2 - \eta \kappa_P} + \sqrt{\theta^4 + 2\eta \theta^2 \kappa_P + 2\eta^2 \kappa_P^2}}{\sqrt{2\eta}} \sim \Omega^2.$$ \(49\)

Expanding around large $\theta$, we find the typical results

$$v_g \approx v_p \frac{\Omega^2}{g_P^2(1 - n_V)}$$

$$\Delta\omega \approx \frac{\Omega^2 \kappa_P}{2g_P^2(1 - n_V)}$$

and therefore

$$\frac{v_g}{\Delta\omega} = \frac{2v_p}{\kappa_P},$$ \(51\)

which agrees with the result for the free photon. Consequently, the effective probe photon interaction range

$$L_M = v_p/\kappa_P$$ \(52\)

is unaffected by the formation of dark-state polaritons and the accompanying reduction of the group velocity. Independent of the mixing angle $\theta$ the inverse interaction range thus remains a small parameter suitable for a perturbative expansion. Note that at fixed $g_P$ both the group velocity and line-width of the dark-state polariton can be conveniently tuned by adjusting the Rabi amplitude $\Omega$. We illustrate this by showing a logarithmic density plot of the frequency and momentum resolved number density of polaritons $n_P(\omega, k)$ in Fig. 6, where the increase in group velocity and decay rate with growing $\Omega$ are clearly visible.

In non-equilibrium physics, the Keldysh and retarded component of the GF are not related by the fluctuation-dissipation theorem. Therefore, the distribution function $F$ introduced in Sec. III C becomes an interesting quantity as it measures the strength of the drive that a given degree of freedom experiences, independent of its actual susceptibility. As the atoms and Lindblad operators are assumed to be distributed homogeneously in space, $F_P$ is independent of momentum. In Fig. 7 we illustrate that despite the broad drive by $\kappa_s(\omega)$, the distribution function of the dark-state polariton has a very sharp peak centered around the resonance with the laser on the $|e\rangle - |s\rangle$ transition, where it reaches the maximally possible value $F_P(\Delta_s) = 2\kappa_s/\kappa_P + 1$. 25
Let us now see how the properties of the EIT-polaritons are affected by coupling the state \( |s\rangle \) to \( |d\rangle \) via the laser with Rabi frequency \( \Omega_s \), but still in the absence of \( E \) photons. In this case, the GF \( G^R \) remains exactly computable in the limit of vanishing polariton density, however now the polarizability is given by

\[
G^R_e(\omega) = \frac{1}{\omega - \Delta_s - \frac{\Omega_s^2}{\Delta_s - \Delta_d + i\gamma_d/2} + i\gamma_e/2}.
\]  

(53)

Since the admixture of \( |d\rangle \) to \( |s\rangle \) introduces losses \( \gamma_{\text{eff}} \approx \Omega_s^2 \gamma_d/(\Delta_d^2 + \gamma_d^2/4) \) to the metastable atomic state – and therefore to the dark-state polariton – without increasing its group velocity, the waveguide is no longer fully transparent. With slow polaritons being mostly atomic it is clear that already a very small Rabi amplitude \( \Omega_s \) drastically increases the opaqueness of the waveguide. This is captured by the suppression of the peak in the distribution function in Fig. 7. Faster and therefore broader EIT polaritons are much less susceptible and thus the maximal value of \( F_P(\omega) - 1 \) once again approaches \( 2\kappa_s/\kappa_P + 1 \) for \( \Omega \to \infty \), whereas it drops to the typically much smaller value \( 2\kappa_s/(4g^2_P(1 - n_V)/\gamma_e + \kappa_P) + 1 \) as \( \Omega \to 0 \) (see Fig. 8).
FIG. 7. Distribution function of the perturbed dark-state polariton in the vicinity of the EIT condition at the same parameters as in Fig. 6 and $\Omega_s = 0.01$. Clearly the occupation of the slower polaritons is more strongly suppressed by the induced losses. For comparison we also added (in grey) the distribution function of the unperturbed EIT polaritons (i.e. $\Omega_s = 0$) for $\Omega = 0.25$.

V. NEXT-TO-LEADING ORDER: INTERACTIONS BETWEEN POLARITONS

The strong dependence of EIT polaritons at large $\theta$ on the properties of the metastable state $|s\rangle$ can be exploited to enhance the effect of interactions. However, one quickly realizes that to leading order in $1/L$, that is to say simultaneously in $1/L_E$ and $1/L_P$, the polaritons cannot interact. Indeed, to order $(1/L)^0$ the only interaction is a Hartree self-energy for the $s$-propagator of the type shown in the last diagram of Fig. 1b). While one can include arbitrarily many Hartree insertions (two in the above figure), as soon as a photon insertion of the type shown in the second diagram of Fig. 1b) appears in an atomic line, it will necessarily induce a suppression by $1/L$. Avoiding this will in particular exclude the appearance of any atomic $g$- or photonic $P$-propagators in self-energies to the $s$-propagator, and therefore prevent us from populating the $|s\rangle$ or the $|d\rangle$ level. The latter are not directly pumped and consequently, without $O(1/L)$-insertions, empty. The distribution functions $F_{s,d}(\omega)$ are thus
FIG. 8. Using the same parameters as in Fig. 7, the EIT window is mostly destroyed for $\Omega \lesssim 0.2$, despite the very weak coupling to the lossy state $|d\rangle$. On the other hand polaritons with $\Omega \gtrsim 1$ are largely unaffected.

identical to one, which means that all particle-hole diagrams, and in particular all Hartree diagrams vanish. This is nothing else than the statement that there can be no interaction between atoms in state $|s\rangle$ if that level is not populated.

Therefore, in our expansion, interactions between polaritons only start to play a role at $O(1/L)$ and the leading order investigated in the last section is indeed a theory of non-interacting polaritons. All the diagrams for the $P$-photon self-energy up to order $1/L$ are shown in Fig. 9. Note that the version of diagram c) with the $E$-propagator substituted by a $P$-propagator has to be excluded according to the Feynman rules discussed in section III D. In general, the order of a diagram is given by $(1/L)^n$, where $n$ is the number of total loops minus the number of atomic loops.

The fact that interactions take place at higher loop-order is a generic feature of polaritons formed by hybridizing probe photons with internal atomic excitations: If the atoms are initialized in the ground state and only probe photons are capable of exciting this initial configuration, then one will first need to populate the interacting atomic level, before atoms
FIG. 9. All contributions to the polariton self-energy at next to leading order in $1/L$. The bold lines for the probe photon indicate that all powers of the leading order polarization bubble (see Fig. 4) have to be inserted as well. For the purpose of clarity, we are not specifying the atomic states and also not using the loop-reduction simplification illustrated in section III E.

- and thus polaritons – can interact.

VI. THE LIMIT OF LONG-RANGED ATOM-ATOM INTERACTIONS

We will begin our discussion of interactions between polaritons with the limit of infinitely ranged exchange photons ($L_E \to \infty$), which implies infinitely ranged atom-atom interactions. In this case all diagrams can be resummed completely, resulting in a fully controlled field theory of a non-equilibrium system with strong light matter interactions. In this case, no further assumptions regarding $L_P$ are required. In particular, we are allowed to enter the regime of large single-atom cooperativities with respect to the propagating photons. We shall see that new phases emerge and that the corresponding phase transitions can be described in a quantitative manner. Before presenting the full theory in the $L_E \to \infty$ limit, we first demonstrate a simplified version of the diagrammatics that allows for an enhanced polariton density stabilized by dissipative interactions, which is at the core of the emergent new phases.
A. Reduced theory for dissipatively-interacting polaritons

In the present section we will consider only a particular subclass of the next-to-leading order interactions which does not involve momentum transfer between photons or equivalently between polaritons. The only diagram of this subclass contributing to the \( P \)-photon self-energy is shown in Fig. 9a). We will see that such a Hartree-like term can have very interesting effects on the polariton transparency window and induce a phase transition in the steady state. Importantly, while this reduced set of diagrams will not typically yield quantitative results, it helps to illustrate many useful physical concepts and provides a simple application of the techniques outlined in Sec. III. We therefore employ it as an instructive introduction into the theory of strongly interacting polaritons.

1. Self-consistent Dyson equations

We begin with the simultaneous expansion in \( 1/L_E \) and \( 1/L_P \), which in next to leading order results in the diagrams shown in Fig. 9. Of these diagrams a) and d) are suppressed by \( 1/L_P \), c) is proportional to \( 1/L_E \) and b) depends on a combination of both lengths that approaches \( 1/\max(L_E, L_P) \) if both length scales differ a lot. Consequently, with \( L_E \to \infty \) only diagrams 9a) and d) need to be considered. In a perturbative expansion, that is if the single atom cooperativity \( C_P = g_P^2/(\kappa_P \gamma e L_P) \ll 1 \) no self-consistent treatment, apart from the resummation of all RPA diagrams that give rise to EIT, is required. At the same time, these weak interactions only slightly perturb the bare EIT and no qualitatively new effects are encountered as these require coupling strengths that are large enough to compensate for the bare \( |s\rangle \to |d\rangle \) coupling \( \Omega_s \), thereby breaking the strict confines of the \( 1/L_P \)-expansion (see Sec. VIA 3). We therefore extend our analysis to strong single atom cooperativities, where all diagrams of the same class as 9a) and d) have to be taken into account. As this becomes somewhat involved, we will introduce the idea of the self-consistent resummation of a class of diagrams and the resulting physical consequences first by using only the diagram in 9a). With all couplings to external lasers made explicit, this diagram takes the form shown in Fig. 10. Clearly, every \( |s\rangle \to |d\rangle \) transition can either be directly driven by a laser acting on a single atom, as is the case in the second transition in the lower loop in Fig. 10 or by the exchange of an \( E \)-photon with another atom that in turn couples to the laser, which is
realized for the excitation from $|s\rangle$ to $|d\rangle$ in the lower loop of Fig. 10. The interchangeability of the single- and multiple-atom processes gives rise to an infinite set of diagrams that is conveniently captured by a self-consistent treatment of the skeleton diagram.

The resulting approximation is depicted diagrammatically in Fig. 11. As we explain in the following, the corresponding self-consistent Dyson equations can be simplified such that they require finding only a single number $\chi$ as the solution of a nonlinear integral equation.

In close analogy to the formalism of Sec. IV the probe photon GFs are dressed by excitations
FIG. 11. Diagrammatic representation of the Dyson equations in the reduced Hartree-like approximation. The latter neglects all interaction diagrams at next-to-leading order except that in Fig. 10a). For the purpose of clarity, we refrain from using the loop-reduction simplification introduced in section III E.

induced in the medium. The result

$$G^R_P(\omega, k) = \left[ G^A_P(\omega) \right]^* = \frac{1}{\omega - \Delta_P(k) - \Sigma_P^R(\omega, k) + i\kappa_P/2}$$

$$G^K_P(\omega, k) = G^R_P(\omega, k) \left( \Sigma^K_P(\omega, k) - i\kappa_P - 2i\kappa_s \right) G^A_P(\omega, k) \quad (54)$$
is therefore still fully determined by the polarization bubble, which using the Kramers-Kronig relations can again be put in the closed form

\[ \Sigma^R_P(\omega, k) = \frac{g_P^2(1 - n_V)}{\omega - \Omega^2 G^R_s(\omega) + i\gamma_e/2} \]

\[ \Sigma^K_P(\omega, k) = 2i\text{Im} \Sigma^R_P(\omega, k). \] (55)

However now the propagator of state \( |s\rangle \)

\[ G^R_s(\omega) = \left[ G^A_s(\omega) \right]^* = \frac{1}{\omega - \Delta_s - \Sigma^R_s(\omega) + i\epsilon/2} \] (56)

has a modified coupling to state \( |d\rangle \):

\[ \Sigma^R_s(\omega) = \frac{\left( \Omega_{s}^{\text{eff}} \right)^2}{\omega - \Delta_d - \Delta_s + i\gamma_d/2}, \] (57)

where \( \Omega_{s}^{\text{eff}} = \Omega_s |1 + \chi| \) includes the effects of the direct coupling rate \( \Omega_s \) as well as those due to the interactions. Here \( \chi \) is simply a complex number, which stems from the fact that the exchange photon mediating the interaction between different polaritons carries zero momentum and – in the rotating frame – zero frequency as well.

In the polarization bubbles of the exchange photon the non-linear Feynman rules forbid a dressing of \( G_s \) by \( |d\rangle \), which thus requires the definition of a second type of \( s \)-propagator

\[ G^R_{\not{s}}(\omega) = \left[ G^A_{\not{s}}(\omega) \right]^* = \frac{1}{\omega - \Delta_s - \Sigma^R_{\not{s}}(\omega) + i\epsilon/2} \] (58)

that couples exclusively to \( |e\rangle \), which in turn can emit and reabsorb a probe photon. This is accounted for by defining

\[ \Sigma^R_{\not{s}}(\omega) = \frac{\Omega^2}{\omega - \Sigma^R_{\not{e}}(\omega) + i\gamma_e/2} \] (59)

and

\[ \Sigma^K_{\not{s}}(\omega) = 2i\text{Im} \Sigma^R_{\not{s}}(\omega) + \delta \Sigma^K_{\not{s}}(\omega) \]

\[ \delta \Sigma^K_{\not{s}}(\omega) = \frac{\Omega^2 \left( \Sigma^R_{\not{e}}(\omega) - 2i\text{Im} \Sigma^R_{\not{s}}(\omega) \right)}{(\omega - \text{Re} \Sigma^R_{\not{e}}(\omega))^2 + (\gamma_e/2 - \text{Im} \Sigma^R_{\not{e}}(\omega))^2}. \] (60)

Here the self-consistency loop closes, as the self-energy \( \Sigma^R_{\not{e}} \) depends on the probe photon propagator via

\[ \Sigma^R_{\not{e}}(\omega) = \frac{i}{2} g^2 \int_{-\infty}^{\infty} d\omega' \int_{-\pi}^{\pi} \frac{dk}{2\pi} G^R_P(\omega - \omega', k)G^R_{\not{s}}(\omega') + G^R_P(\omega - \omega', k)G^R_{\not{s}}(\omega') \] (61)
\[ \delta \Sigma^K_e(\omega) = \Sigma^K_e(\omega) - 2i \Im \Sigma^K_e(\omega) = \frac{i}{2} g_p^2 \int_0^\infty \frac{d\omega'}{2\pi} \int_{-\pi}^\pi \frac{dk}{2\pi} \delta G^R_p(\omega - \omega', k) \delta G^K_g(\omega'), \]  

(62)

As announced at the beginning of this section, the self-consistent functional equations \( G^R_{P,\star} = G^R_p [G^R_{P,\star}, G^K_{P,\star}] \) and \( G^K_{P,\star} = G^K_p [G^R_{P,\star}, G^K_{P,\star}] \) have been reduced to a single parameter satisfying a fixed point equation \( \chi = \chi(\chi_{\star}) \). As mentioned before, this is in part due to the Hartree nature of the interactions considered here, which implies that the functional form of \( \Sigma^R_R \) is fixed and analytically known. On the other hand it is a consequence of the non-linear Feynman rules, which enforce an unoccupied propagator \( G^K_s \) and therefore \( \Sigma^K_s = 2i \Im \Sigma^K_R \), which reduces the number of coupled equations.

The frequency integral in the first of the two expressions in Eq. (61) is trivial, as \( G^K_s(\omega) \propto \delta(\omega) \).

Since the poles of \( G^K_p(\omega) \) can be found analytically, also the frequency integral in the second term of \( \Sigma_e(\omega) \) can be solved exactly via the residue theorem, such that only the momentum integration has to be evaluated numerically. After application of the residue theorem one obtains

\[ \Sigma^K_e(\omega) = 2i \Im \Sigma^K_e(\omega) - i \kappa_s \int \frac{dk}{2\pi} g_p^2 (2 - 2n_V) G^R_p(\omega + i\epsilon/2, k), \]  

(63)

where \( n \in \{1, 2, 3, 4\} \), \( \omega_n(k) \) are the poles of \( G^K_p(\omega, k) \) and

\[ f(\omega) = (\omega + i\gamma_e/2)(\omega - \Delta_e + i\epsilon/2)(\omega - \Delta_s - \Delta_d + i\gamma_d/2) 
- \Omega^2(\omega - \Delta_e - \Delta_d + i\gamma_d/2) - \left( \Omega_{\alpha}^{\text{eff}} \right)^2(\omega + i\gamma_e/2). \]  

(64)

With all GFs depending solely on the parameter \( \chi \), we are left with the task to solve for it self-consistently. The corresponding equation can again be read off from Fig. 11 and states

\[ \chi = \frac{\Sigma^R_E(0)}{\Delta_E(0) - \Sigma^R_E(0) + i\kappa_E/2}. \]  

(65)

So far, there is no ambiguity regarding the non-linear Feynman rules. In the polarization bubbles of the exchange photon however, these partially forbid dressing the propagator of state \(|d\rangle \) via couplings to the metastable state. Employing the strict interpretation where
\( G_d^R \) remains undressed, the exchange photon self-energy reads

\[
\Sigma^R_E(\omega) = \frac{i}{2} \int \frac{d\omega'}{2\pi} g_E^2 G^R_E(\omega') G^A_E(\omega') \delta \Sigma^K_e(\omega') G_d^R(\omega + \omega')
\]

(66)

with

\[
G_d^R(\omega) = \frac{1}{\omega - \Delta_s - \Delta_d + i\gamma_d/2},
\]

(67)

If on the other hand the lenient rule is applied one is to use

\[
G_d^R(\omega) = \left( [G^R_{d0}]^{-1}(\omega) - \frac{(\Omega_{\text{eff}}^s)^2}{\omega - \Delta_s - \frac{\Omega^2}{\omega + i\gamma_s/2}} \right)^{-1},
\]

(68)

which includes all possible admixtures of atomic states to \(|d\rangle\), as the insertion of the ground-state can always be excluded by the methods introduced in Sec. III D. Furthermore, \(\Sigma^K_R\) is to be complemented by

\[
\Sigma^K_R \rightarrow \Sigma^K_R + \frac{(\Omega_{\text{eff}}^s)^2}{\omega - \Delta_s - \Delta_d + i\gamma_d/2},
\]

(69)

with the dependence of \(\delta \Sigma^K_e\) on \(\Sigma^K_R\) and \(\delta \Sigma^K_e\) remaining unaffected.

Choosing among these two ways of applying Feynman rules affects the propagation of the exchange photons and hence the light-mediated atom-atom interactions. The photon propagator is ultimately given by

\[
G^R_E(\omega, k) = \left( G^A_E(\omega) \right)^* = \frac{1}{\omega - \omega_E(k) - \Sigma^R_E(\omega, k) + i\kappa_E/2}
\]

\[
G^K_E(\omega, k) = G^R_E(\omega, k) \left( \Sigma^K_E(\omega, k) - i\kappa_E \right) G^A_E(\omega, k),
\]

(70)

with

\[
\Sigma^K_E(\omega, k) = \Sigma^K_0(\omega, k) = 2i\text{Im}\Sigma^R_E(\omega, k).
\]

(71)

Interestingly, the phase of \(\chi\) can be adjusted via the detuning between the band-edge of the exchange photon and the laser \(\Omega_s\). Its amplitude depends on the density of atoms in the metastable state \(n_s\) and on the coupling constants, giving a great deal of control over the type and strength of backaction to be realized.

For numerical purposes, iterating equations (54) through (71) having previously initialized...
the system with some \( \Omega_{\text{eff}} = \Omega_s \) is immensely inefficient, as convergence will fail when approaching a phase-transition\(^{10}\). We avoid this problem by instead fixing \( \Omega_{\text{eff}} \) and determining \( \Omega_s(\Omega_{\text{eff}}, \chi) \), which requires no iterations at all. This actually means that the value of \( \Omega_s \) corresponding to the solution is not known a priori. However, for the computation of the entire phase-diagram this does not matter as eventually a result for any value of \( \Omega_s \) will have been produced.

2. Results: Non-equilibrium phase transition of the transparency window

![Flow diagram of the effective relative coupling strength |1 + \chi| as a function of the externally adjustable parameter \( \Omega_s \), where otherwise the same parameters as in Fig. 6 are used together with the lenient interpretation of the non-linear Feynman rules as well as \( \Delta_E(k = 0) = -1 \), \( \kappa_E = 5 \) and \( g_E = 10 \).](image1)

![The same diagram as in Fig. 12 but using the strict version of the non-linear Feynman rules.](image2)

A particularly interesting question which can be addressed with this newly developed
formalism is whether the transparency window can be restored by interaction-related effects. If this was not the case one would have to limit the system to weak interactions or accept that strongly interacting polaritons in photonic crystal waveguides are necessarily very lossy. If on the other hand such a restoration of the transparency window is possible, it requires a condition similar in spirit to that of the original EIT, however with destructive interferences between the laser and the exchange photon that drastically reduce the coupling to state $|d\rangle$. As it turns out, such interferences are indeed predicted within our approach and involve the last four diagrams in Fig. 11).

This many-body phenomenon, which can be named “interaction-induced transparency” as opposed to the standard single-particle “electromagnetically-induced transparency”, is analyzed elsewhere[27], also in relation to its observability for realistic experimental parameters in the context of PCW and tapered fibers. In the remainder of this section, we provide a complementary analysis focusing on the nature of the underlying non-equilibrium phase transition and discuss the fundamental mechanism from a more formal perspective as an application of our diagrammatic approach.

The reconstruction of the transparency window can be attributed to the positive feedback brought about by the dependence of $\chi$ on the excitation density: $\chi \propto n_s$, which stabilizes both a low density i.e. opaque phase and a high density i.e. transparent phase, separated by a first-order phase transition. The mechanism behind this can be understood by studying Figs. 12 and 13, which show the amplitude and sign of the variation in the flow of the quantity $|1 + \chi|$ during the evaluation of the self-consistence equation (65). If the system is initialized with a certain value of $\chi$ such that $\delta|1 + \chi|$ is positive, the system will flow towards the opaque phase and vice versa, if $\delta|1 + \chi| < 0$, the system is unstable towards the transparent phase. Consequently, only those parameter combinations with $\delta|1 + \chi| = 0$ and a negative slope in $\delta|1 + \chi|$ as a function of $|1 + \chi|$ are stable and therefore marked with a red line in Figs. 12 and 13. In sufficiently strongly driven systems we witness the emergence of a bistability: for a given Rabi amplitude $\Omega_s$ two stable solutions exist. They differ significantly in the effective coupling $\Omega_s^{\text{eff}}$ and in the occupation of dark-state polaritons. Quite surprisingly we find a stable transparent solution with $\Omega_s^{\text{eff}} \ll \Omega_s$, which entails significantly reduced losses compared to the non-interacting case with $g_E = 0$. Remarkably, the stable ratio $\Omega_s^{\text{eff}} / \Omega_s$ is smallest for purely dissipative interactions, that is, when $\Sigma_E R(0)$ is purely imaginary. In this case, the phase shift between the $E$-photon-mediated driving of
the $s-d$ transition and the direct driving via $\Omega_s$ is the most destructive. This results in small losses for the dark-state polaritons, at least if there are enough to create a sufficiently large backaction in the form of $\Sigma^R_E(0)$. A comparison between Figs. 12 and 13 demonstrates that for these rather small values of $\Omega_s$ the choice of the non-linear Feynman rules does not affect the results appreciably. For the remainder of this section, we will therefore focus on the strict implementation of the Feynman rules.

In combination with the possibility of the simultaneous stability of an opaque and a transparent phase, a first order phase transition similar to that between a gaseous and a liquid phase emerges: above a critical bare laser strength $\Omega_{s_c}$ an increasingly strong hysteresis is observed as the source intensity $\kappa_s$ is increased (see Fig. 14). However, at exactly the critical laser strength, the first order phase transition ends in a critical point, where the phase transition is continuous and of mean-field type. This is to be expected by a Hartree-type theory with infinitely ranged interactions and we verify this by fitting the numerical data for $n_s(\kappa_s, \Omega_{s_c})$ with a power law and extracting the critical exponent $\delta = 3 \pm 0.01$, consistent with the Ising universality class (see Fig. 15).

We note that in the regime of the first order phase transition, the difference in polariton

![FIG. 14. Hysteresis of the polariton density, evidenced in $n_s$ for scans at the incoherent drive strength $\kappa_s$ for different values of $\Omega_s$. In panel a) the system is initialized in the opaque phase with $\kappa_s = 0$, whereas panel b) uses $\kappa_s = 2$ in the transparent phase as a starting point. Below the critical Rabi amplitude $\Omega_{s_c} \approx 0.0851$ both scans are identical. However above $\Omega_{s_c}$ the initial phase is stabilized against fluctuations induced by slow scans and a hysteresis curve becomes observable. The parameters used are the same as in Fig. 12.](image-url)
FIG. 15. Power law fit to the density of excited atoms at the critical coupling strength $\Omega_{s_c} \approx 0.0851$ for the same parameters as in Fig. 12. The critical exponent of the order parameter as a function of the drive strength is determined to be $\delta = 3 \pm 0.01$.

density between the opaque and transparent solution is typically large. This can be seen from the distribution function (see Fig. 16) as well as from the frequency- and momentum-resolved photonic number density of Fig. 17. One thus concludes that, far away from the critical point in the opaque phase the system behaves essentially as a non-interacting theory: the occupation numbers are so small that interactions via exchange photons play no role and the bare – but due to $\Omega_s$, lossy – EIT is recovered.

In the transparent phase on the other hand an only weakly perturbed $\Lambda$-scheme is restored, which seems to imply that the effective degrees of freedom are again only weakly interacting. Correspondingly, many simple correlation functions can be described by an effective free theory. However, except for the limit of vanishing $\Omega_s$, the response of the system to external perturbations will be very different compared to the free theory discussed in Sec. IV.
3. Analytic estimates and requirements of the bistable regime

Due to the simplicity of the reduced theory presented in this section, we can actually give some analytic estimates for the conditions necessary for a phase transition. Due to the typically large atomic admixture $\theta$ to the dark-state polaritons, even for relatively strong driving $\kappa_s \sim \kappa_P$, a slow group velocity gives rise to only a small photon number density

$$n_P = i \int \frac{d\omega}{4\pi} \int \frac{dk}{2\pi} \delta G_P^K(\omega, k) < \theta n_s \ll 1.$$  \hspace{1cm} (72)$$

Here the first inequality results from the fact that only photons in a narrow frequency interval actually form dark-state polaritons. Most photons instead hybridize into bright polaritons, that involve the decaying excited atomic states, resulting in even smaller occupations.

Of the two contributions to $\Sigma^R_\nu$ in (63), the second one thus dominates. Typically, in PCW or tapered fibers, the photonic bandwidth is several orders of magnitude larger than the inverse life times of all atomic states. It is therefore well justified to approximate the photon
FIG. 17. The frequency and momentum resolved photon number density $n_P(\omega, k)$ in the transparent phase (left) exhibits an almost perfect transparency window, whereas the opaque phase (right) with the same parameters shows strongly dissipative polaritons (note the difference by almost three orders of magnitude in the maximal spectral density). The almost flat blue line corresponds to the atomic level $|d\rangle$, that hybridizes with the probe photon to form a fourth, bright polariton branch. For small values of $\Omega_s$, which is the case for the parameters of Fig. 12, which are also at use here, this hybridization remains weak and the new polariton branch consequently is essentially unoccupied.

spectrum as linear. We do so by writing their retarded GF as a sum of left- and right-movers

$$G_P^R(\omega) = \frac{1}{\omega - \Delta_P^{(0)} - v_P k - \Sigma_P^R(\omega) + i\kappa_P^P} + \frac{1}{\omega - \Delta_P^{(0)} + v_P k - \Sigma_P^R(\omega) + i\kappa_P^P}. \quad (73)$$

For $L_P = \kappa_P/v_P \gg 1$ the EIT window in momentum space is much narrower than the inverse lattice constant $1/a$ and thus far away from the band edge a linearized spectrum suffices to reproduce the results obtained from any Bloch wave with the same group velocity in the EIT window.

Together with the observation that, since the atoms are fixed in space, $\Sigma_P^R(\omega)$ is momentum independent, this allows to find

$$\Sigma_P^R \approx -ig_P^2(2 - n_V)|u_k^P(0)|^2/v_P$$

and

$$\delta\Sigma_P^K \approx -g_P^2(1 - n_V) \frac{\kappa_s}{v_P \kappa_P^P/2 - 3\Sigma_P^R(\omega)}, \quad (74)$$

where the momentum integral has been approximated by an integral along the entire real axis. This result can be used to approximate the number density of atoms in the metastable
state by
\[ n_s = i \int_{-\infty}^{\infty} \frac{d\omega}{4\pi} |G_R^s(\omega)|^2 \delta \Sigma_K^R(\omega) = i \int_{-\infty}^{\infty} \frac{d\omega}{4\pi} |G_R^R(\omega)|^2 |G_e^R(\omega)|^2 \delta \Sigma_e^K(\omega) \]
\[ \approx \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \kappa_s(\omega) \left| \frac{1}{(\omega - \Delta_s) + i\gamma_e} \left( \omega + (2 - n_V)\frac{g_P^2}{v_P} + i\gamma_e/2 \right) - 1 \right|^2 \frac{g_P^2}{\Omega^2 v_P (\kappa_P/2 - 3 \Sigma_R^E(\omega))}. \]  

(75)

As can be extracted from Figs. 12 and 13, the system becomes bistable once
\[ 0 > \frac{d\Omega_s}{d\Omega_{\text{eff}}} = \frac{d}{d\Omega_{\text{eff}}} \frac{\Omega_{\text{eff}}^s}{1 + \chi(\Omega_{\text{eff}})} \]  

which, using the explicit form (65) can be rewritten as
\[ \frac{d\chi}{d\Omega_{\text{eff}}^s} > \left| \frac{-\Delta_E(0) + i\kappa_E/2}{\Omega_{\text{eff}}^s (-\Delta_E(0) - \Sigma_R^E(0) + i\kappa_E/2)} \right|. \]  

(77)

In the ideal case of a resonance between the exchange photon and the corresponding laser \((\Delta_E(0) = 0)\) as well as strong coupling \(g_E\), such that \(|\Sigma_R^E(0)| \gg \kappa_E\), this still requires
\[ \frac{dn_s \Omega_{\text{eff}}}{d\Omega_{\text{eff}}^s} < 0. \]  

(78)

A condition, that can be satisfied only if
\[ 3 \Sigma_P^R(0) - \Omega_{\text{eff}} d \Sigma_P^R(0) > \kappa_P/2, \]  

(79)

where we used (75) with the absolute value approximated by unity as an upper bound. Since the minimum of the frequency dependent loss rate
\[ -3 \Sigma_R^R(\omega) \approx \sigma + \eta(\omega - \Delta_s)^2 \quad \text{with} \quad \sigma = \frac{2(\Omega_{\text{eff}}^s)^2 g_P^2 (1 - n_V)|u_P^R(0)|^2}{\gamma_e (\Omega_{\text{eff}}^s)^2 + \gamma_d \Omega^2} \quad \text{and} \quad \gamma_d = \frac{\gamma_d^2 + 4 \Delta_d^2}{\gamma_d}, \]  

(80)

for slow polaritons is tightly focused around \(\omega = \Delta_s\), this is a reasonably good approximation. Using the just stated expansion of the probe photon self-energy around \(\Delta_s\), one finds the left hand side of Eq. (79) to be maximized for
\[ \Omega_{\text{eff}}^s = \sqrt{\frac{\gamma_d \Omega^2}{3\gamma_e}}, \]  

(81)

where one finds a strong collective coupling satisfying
\[ g_P^2 > 2 \gamma_e \kappa_P. \]  

(82)
or equivalently a large collective cooperativity $L_P C_P > 2$ to be a necessary condition for the emergence of a bistability. While, due to the rough approximations used here, this is only a lower bound on the collective cooperativity, it clearly shows that the type of phase transition discussed here is not amenable to a purely perturbative approach.

Instead of calculating a lower bound for the collective cooperativity $C_P$ we can also search for a rough estimate that includes all relevant scales. To do so, we approximate $n_s \approx p C_P \Omega_s^2 / \Omega^2$, where $p = 2 \kappa_s(0) / \kappa_P$ is the pump ratio, indicating how strongly the probe photons near the EIT condition are driven compared to their losses. Inserting this expression for $n_s$ into $|\Im \Sigma_R E(0)| \gtrsim \kappa_E$, which is necessary for a highly non-perturbative regime, yields the final strong coupling condition

$$p L_E C_E C_P \Omega_s^2 / \Omega^2 \gtrsim 1. \quad (83)$$

An actual bistability additionally requires an efficient backaction of the losses in the dressed state $|s\rangle$ onto the dark-state polariton density. Therefore, typical systems that exhibit a phase transition satisfy Eq. (83) by more than one order of magnitude. For example, for the parameters of the critical point in Fig. 15, one has $p L_E C_E C_P \Omega_s^2 / \Omega^2 \approx 58$.

### B. Quantitative theory in the infinite-range limit

The reduced class of diagrams discussed in the previous section is helpful to obtain a general idea about the emergence of a phase transition between the two limits of a perfectly restored transparency window deep within the transparent phase on the one hand, and an empty system in the opaque phase on the other hand. Our main goal, however, is the quantitative description that extends all the way to the critical point and the bistable region. In order to achieve this, one has to include all diagrams that can be created self-consistently from the two diagrams in Fig. 9a) and d). The resulting theory is illustrated in terms of Feynman diagrams in Fig. 18, which differs from the reduced theory of the previous section by the addition of the Fock diagram to the Dyson equation of the exchange photon (see last diagram in the third line of Fig. 18). Note that at this level of the theory the exchange photon obtains a Nambu structure, which requires us to extend the Kramers-Kronig relations of Sec. III C to anomalous GFs, which we will do in the following.
FIG. 18. Complete diagrammatics in the limit $L_E \to \infty$. Note that we do not show the anomalous components of the self-energy for the E-photon. Those are however obtained from the last diagram in the second line upon an exchange of laser and E-photons acting on the same transition. We included the anomalous components in the calculation (see section VI B 1).

1. **Anomalous Green’s functions**

The appearance of anomalous GFs for the exchange photons is caused by the presence of a background coherent contribution to the $E$-photon field. This has to be expected from the theory presented in Sec. VIA as an E-photon can be resonantly excited by the external laser field with strength $\Omega_s$. The net effect is that a coherent component i.e. a condensate arises in the connected part of the two-point function of the exchange photon as an addition to the background coherent laser field. This effect is described by the last diagram in the third line of Fig. 18 where at each atom the order in which the $E$-photon and the laser
are coupled to a given transition can be chosen arbitrarily. While this formally creates anomalous contributions to the self-energy, it underlines the physical argument that from the perspective of fixed atoms exchange photons and laser photons are indistinguishable if their frequency is identical.

It is important to understand that this condensation of $E$-photons will not be inherited by the probe photons. Consequently, no polariton condensate can be generated unless the probe photons themselves are directly coupled to an inverted bath (which we excluded from the outset). Similar to what is done in equilibrium, each component of the non-equilibrium GF can be augmented by a Nambu structure. For the retarded and the Keldysh part of the GF one typically defines the following $2 \times 2$ matrix

$$G^R(\omega, p) = -i \begin{pmatrix} \langle a^{\text{cl}}(\omega, p)\bar{a}(\omega, p) \rangle & \langle a^{\text{cl}}(\omega, p)a^q(-\omega, -p) \rangle \\ \langle \bar{a}^{\text{cl}}(-\omega, -p)\bar{a}(\omega, p) \rangle & \langle \bar{a}^{\text{cl}}(-\omega, -p)a^q(-\omega, -p) \rangle \end{pmatrix}$$

and the same for $G^K(\omega, p)$ with all quantum fields (index $q$) replaced by classical fields (index $\text{cl}$).

The diagonal entries then describe ordinary GFs, while the off-diagonal, so-called anomalous components, are non-zero only in the presence of a condensate. For the retarded GF the Dyson equation takes the same form as in equilibrium

$$G^R = G^R_0 + G^R_0 \cdot \Sigma \cdot G^R,$$

where $G^R_0$ is purely diagonal. Retarded, advanced and Keldysh GFs are once again not independent and one finds relations between them which are very similar to the case where anomalous GFs are absent. The retarded and advanced components have namely to satisfy $G_{\sigma p}^R(\omega, p) = G_{\bar{\sigma}p}^A(-\omega, -p)$, where $\sigma, p \in \{1, 2\}$ and $\bar{\sigma}$ is the complement of $\sigma$. The proof follows from a direct inspection of the respective Feynman diagrams: exchanging the external legs reverses the direction of the momentum and energy flow and simultaneously reverses causality, thereby equating the off-diagonal entries of $G^R$ and $G^A$. Additionally $[G^A]^\dagger = G^R$ follows immediately once the definition is evaluated on the Keldysh contour. Therefore, the advanced GF never has to be calculated and the retarded GF can be restricted to only two independent functions:

$$G^R(\omega, p) = \begin{pmatrix} G^R_{11}(\omega, p) & G^R_{12}(\omega, p) \\ [G^R_{12}]^* (-\omega, -p) & [G^R_{11}]^* (-\omega, -p) \end{pmatrix}.$$
Furthermore, considering all self-energy diagrams order-by-order, one can prove that 
\[ G_{\sigma,\sigma}^{R}(\omega, p) = G_{\bar{\sigma},\sigma}^{R}(\omega, p), \] 
which is true for any uniform bosonic system in and out of equilibrium\(^{48}\).

Taking the conjugate transpose of the Keldysh component can be done immediately in frequency space and a comparison of elements reveals the anti-hermitian structure, i.e. \[ [G^{K}]^\dagger = -G^{K}. \] We can thus still use the parametrization \[ G^{K} = G^{R} \cdot F - F \cdot G^{A}, \] with a hermitian matrix \( F \). The fact that \( F = \sigma_z \) for an empty system accounts for the reversed order of operators between the components of the first and those of the second row of \( G^{K} \). Exchanging incoming and outgoing particles in \( G^{K} \) furthermore allows to identify 
\[ G_{\sigma,\rho}^{K}(\omega, p) = -[G_{\bar{\sigma},\bar{\rho}}^{K}]^\ast(\omega, -p). \] The Dyson equation for the Keldysh component directly generalizes to the Nambu structure: 
\[ G^{K} = G^{R} \cdot (\Sigma^{K} - D_{0}^{K}) \cdot G^{A}, \] which together with the other symmetries implies

\[ \Sigma^{K}\sigma(\omega, p) = 2 \begin{pmatrix} i\Im \Sigma_{11}^{K}(\omega, p) & \Re \Sigma_{12}^{K}(\omega, p) \\ -\Re \Sigma_{12}^{K}(\omega, -p) & i\Im \Sigma_{11}^{K}(\omega, -p) \end{pmatrix} \] (87)

for the empty system and

\[ \delta \Sigma^{K}(\omega, p) = \Sigma^{K}(\omega, p) - \Sigma^{K}\sigma(\omega, p) = \begin{pmatrix} \delta \Sigma_{11}^{K}(\omega, p) & \delta \Sigma_{12}^{K}(\omega, p) \\ -[\delta \Sigma_{12}^{K}]^\ast(\omega, p) & \delta \Sigma_{11}^{K}(\omega, -p) \end{pmatrix} \] (88)

for excitations above the vacuum. With these definitions the Kramers-Kronig relations of section III C remain valid without limitation. For simplicity we only provide the Kramers-Kronig relations for one-dimensional convolutions with some normal GF labeled \( G_2 \), keeping in mind that higher dimensional generalizations take exactly the same form:

\[ \int \frac{d\omega'}{2\pi} \left( G_{\sigma,\rho}^{K}(\omega, \omega')G_{2}^{R}(\omega - \omega') - G_{\bar{\sigma},\rho}^{R}(\omega, \omega')G_{2}^{K}(\omega - \omega') \right) = -2\delta_{\rho,2} \int \frac{d\omega'}{2\pi} \left( G_{\sigma,\rho}^{R}(\omega, \omega')G_{2}^{R}(\omega - \omega') - G_{\bar{\sigma},\rho}^{A}(\omega, \omega')G_{2}^{A}(\omega - \omega') \right), \] (89)

as well as

\[ \int \frac{d\omega'}{2\pi} \left( G_{\sigma,\rho}^{K}(\omega, \omega')G_{2}^{K}(\omega - \omega') - G_{\bar{\sigma},\rho}^{R}(\omega, \omega')G_{2}^{R}(\omega - \omega') - G_{\sigma,\rho}^{A}(\omega, \omega')G_{2}^{A}(\omega - \omega') \right) \]
\[ = -2 \int \frac{d\omega'}{2\pi} \left( \delta_{\rho,2}G_{\sigma,\rho}^{R}(\omega, \omega')G_{2}^{R}(\omega - \omega') + \delta_{\sigma,2}G_{\sigma,\rho}^{A}(\omega, \omega')G_{2}^{A}(\omega - \omega') \right). \] (90)

Note that due to the symmetries of the diagonal entries of the GFs, these two relations already fully incorporate the four equations derived in section III C.
2. Dyson equations

Having introduced the anomalous non-equilibrium GFs we can now solve the self-consistent Dyson equations shown in Fig. 18, where in order to simplify the notation we have introduced the matrix GF $G_{sd}$ for the states $|s\rangle$ and $|d\rangle$. In absence of any diagrams of order $1/L_E$, it is fully determined by the corresponding submatrix of $[G_{a,0}^R]^{-1}$ (see Eq. (19)), but with the effective Rabi amplitude $\Omega_{s}^{\text{eff}} = \Omega_s |1 + \chi|:

$$
G_{sd}^{R/K} = \begin{pmatrix} G_{ss}^{R/K} & G_{sd}^{R/K} \\ G_{ds}^{R/K} & G_{dd}^{R/K} \end{pmatrix} = \begin{pmatrix} \omega - \Delta_s + i\epsilon/2 & -\Omega_{s}^{\text{eff}} \\ -\Omega_{s}^{\text{eff}} & \omega - \Delta_d - \Delta_s + i\gamma_d/2 \end{pmatrix}^{-1}.
$$

(91)

In fact, as indicated by the last line in Fig. 18 and in analogy to Sec. VIA, $\Omega_s$ has to be replaced everywhere by $\Omega_{s}^{\text{eff}}$ and $G_{ss}^{R/K}$ supersedes the identical expression $G_{s}^{R/K}$ used in Sec. VIA. Apart from these notational remarks, the only physical difference between the present theory and the one discussed in section VIA is in the propagator of the exchange photon, which acquires a new self-energy contribution $\Sigma_{E}^{R2}$:

$$
G_{E}^{R} = \left( [G_{E}^{R0}]^{-1} - \Sigma_{E}^{R1} - \Sigma_{E}^{R2} \right)^{-1}.
$$

(92)

While the first term $\Sigma_{E}^{R1}(\omega)$ remains exactly the same as Eq. 66, the second, due to the Nambu structure takes the lengthy form

$$
\Sigma_{E}^{R2}(\omega, k) = \frac{i}{2} g_{R}^{A} g_{E}^{A} \Omega_{s}^{2} \Omega_{s}^{2} |1 + \chi|^2 (1 - n_{V})^2 \int \frac{d\omega'}{2\pi} \frac{dp}{2\pi} [G_{e}^{R}(\omega')G_{ss}^{R}(\omega')^2 |G_{E}^{K}(\omega', p)|^2 \delta G_{P}^{K}(\omega', p) 
$$

$$
\times \left[ [G_{ss}^{R}(\omega + \omega')G_{e}^{R}(\omega + \omega')]^2 G_{P}^{R}(\omega + \omega', p + k) \left( \begin{array}{cc} [G_{d}^{R}(\omega + \omega')]^2 & G_{d}^{A}(\omega')G_{d}^{R}(\omega + \omega') \\ G_{d}^{R}(\omega')G_{d}^{A}(\omega + \omega') & [G_{d}^{R}(\omega')]^2 \end{array} \right) \\ + [G_{e}^{A}(\omega' - \omega)G_{ss}^{A}(\omega' - \omega)]^2 G_{P}^{A}(\omega' - \omega, p - k) \left( \begin{array}{cc} |G_{d}^{R}(\omega')|^2 & G_{d}^{A}(\omega' - \omega)G_{d}^{A}(\omega') \\ G_{d}^{A}(\omega' - \omega)G_{d}^{R}(\omega') & [G_{d}^{A}(\omega' - \omega)]^2 \end{array} \right) \right] \right).
$$

(93)

Some care has to be taken when it comes to determining $\chi$: $\Sigma_{E}^{R2}$ is actually indistinguishable from $\Sigma_{E}^{R1}$ once one of their external legs is substituted with the laser field $\Omega_s$. Consequently, coupling to the coherent field with $\Sigma_{E}^{R2}$ would overcount the diagrams in the last line of Fig. 18. Therefore, $\chi$ is given by

$$
\chi = \sum_{j} \Sigma_{E}^{R1}_{E_{j}j} G_{E_{j}j}^{R} \bigg|_{k,\omega=0}.
$$

(94)
Note that using the real and positive definition $\Omega^{\text{eff}}_s = \Omega_s |1 + \chi|$ in the anomalous components of the exchange photon Green's function is wrong, since it leads to an incorrect behavior of $G_E$ under a global $U(1)$ transformation. This does not matter, however, since all observables depend only on the gauge invariant $|1 + \chi|^2$, which allows us to simplify our calculations. By fixing the real value $\Omega^{\text{eff}}_s$ one can then directly determine the corresponding experimentally relevant parameter $\Omega_s$. From a computational point of view, this makes for a very cheap calculation, as the two-dimensional convolution in Eq. (93) – which has to be calculated only once – only has to be evaluated at $k = \omega = 0$.

Similar to the previous section, the simplified non-linear Feynman rules are not uniquely defined and we thus again have to choose between the strict and lenient way of implementing the rules in order to estimate the error bounds of the simplified diagrammatics. We do so in the same fashion as before, i.e. for the strict rule we use $G^{A/R}_{d_0}$ in Eqs. (93), and (66). For the lenient version we employ $G^{A/R}_d$ according to Eq. (68) together with the replacement (69) for the very same equations.

Before we proceed to discuss the results obtained from the set of coupled Dyson equations introduced in this section, it is instructive to view these calculations from a more conceptual point of view: despite the potentially large single atom cooperativity experienced by the probe photons, their density is assumed to be small, such that dark-state polaritons in the absence of exchange photons are non-interacting quasi-particles. This is correctly captured by the non-linear Feynman rules, which allow for an exact diagrammatic solution of the Yukawa theory in the $g - e - s - M$ sector. If we now consider the additional coupling to level $d$ and include the $E$-photons, we can eliminate the atomic degrees of freedom to obtain an effective theory for the dressed propagating and exchange photons. Indeed, on the one hand the atomic level structure contains the microscopic details necessary for the formation of polaritons, which within the effective theory is incorporated in the dressed $P$-photons, and on the other hand the atoms serve as interaction vertices between one probe photon and an arbitrary number of exchange photons. While the latter may be strongly dressed with probe photons themselves, there are only two processes for this that are allowed by the atomic vertices, namely those in the third line of Fig. 18. The diagrammatic representation of the effective theory is shown in Fig. 19. We stress that this is completely equivalent to the theory presented in Fig. 18. In the first line of Fig. 19 the free polariton propagator is defined and indicated as a curly-line. In the second line the interaction vertices between the
polariton and the \( E \)-photons are illustrated. Out of these, only the first two are shown but actually and infinite number of \( E \)-lines is allowed in the vertex indicated by the dots in the last line of Fig. 19 where all possible interaction-corrections to the polariton self-energy are shown. Luckily all of these vertices can be conveniently resummed as a geometric series, as we have demonstrated earlier in the derivation of the self-consistent equations. Similarly, all possible contributions to the \( E \)-photon self-energy are shown in the third line. However, as every vertex has to involve exactly one probe photon, the number of diagrams here is limited to two.

\[
\begin{align*}
\chi &= \chi + \chi + \chi + \chi \\
\Sigma_{R}^{E} &= \Sigma_{R}^{E} + \Sigma_{R}^{E} + \Sigma_{R}^{E} + \Sigma_{R}^{E} + \ldots
\end{align*}
\]

FIG. 19. Effective theory of dark-state polaritons in the limit of infinite interaction range, i.e. \( L_{E} \rightarrow \infty \). While there is an infinite set of vertices coupling a single probe photon to an arbitrary number of exchange photons, these are conveniently summed up in the geometric series embedded in \( \Sigma_{R}^{E} \). This effective theory is completely equivalent to the one shown in Fig. 18.

### 3. Quantitative results and validity

With the inclusion of all effects at leading order in \( 1/L_{E} \), \( \chi \) is no longer bounded from below by \( -1 \). In fact, it can achieve arbitrarily small values, which can be understood by a closer examination of the effects of \( \Sigma_{R}^{E} \) in terms of the effective theory in Fig. 19 where it is represented as the last diagram of the third line. Within this framework one immediately realizes, that \( \Sigma_{R}^{E} \) describes in fact a particle-hole excitation of a probe photon. Since,
however, this photon itself is strongly dressed, its distribution $F_P(\omega)$ is sharply peaked. This allows for a resonant reallocation of photons from highly occupied frequencies and momenta towards low-occupation regions, by means of the particle-hole excitations in $\Sigma_{E}^{R2}$. Where this is possible, it will act as a locally inverted environment for $G_E$, thereby effectively driving the exchange photons. Since there is no other diagram to counter this effect, the exchange photon propagator can develop a divergence, resulting in $\chi \to -\infty$, which is unphysical. While in general there is nothing wrong with the inverted bath experienced by the exchange photons, one has to pay attention to its effect on $L_E = \sqrt{\alpha_E/\kappa_E^{\text{eff}}}$. The latter namely vanishes as the divergence in $G_E^R$ is approached. Consequently, diagrams at higher order in $1/L_E$ have to be included and these will in turn prevent the unphysical instability in the exchange photon propagator. We will outline the underlying processes in the next section. Nevertheless, as long as $L_E$ remains large enough, $-\chi$ can still become large without forcing us to include subleading orders in $L_E$. This can happen to such an extent, that it actually overcompensates the bare coupling $\Omega_s$ up to the point where a new, strongly interacting phase emerges. This new phase, which will be referred to as “intermediate phase”, is stable, as evidenced by the flow diagrams 20 and 21, which we show again both for the lenient and the strict implementation of the Feynman rules. As there is hardly any quantitative differences between the two versions, we will in the following focus on the strict rules.

Previously, we presented an argument for the emergence of the bistability, whereby an increase in $\Omega_s^{\text{eff}}$ was met with a sufficiently fast decrease of $n_s$ (and of $\chi$), so that $\Omega_s$ itself was reduced, resulting in a non-unique identification $\Omega_s(\Omega_s^{\text{eff}})$, i.e. a bistability. It is exactly the opposite effect that stabilizes the intermediate phase, whereby for small $\Omega_s$ an increase of $\Omega_s^{\text{eff}}$ increases the efficiency of the drive experienced by $G_E^R$, such that $|\chi|$ grows until this effect is exactly balanced by the effects of increased losses discussed in Sec. VI A. If this happens at $\chi \lesssim -1$ a stable intermediate phase exists.

As can be observed in Fig. 22 where the losses $\gamma_d$ have been increased tenfold compared to Figs. 12 and 13, the stability of the transparent phase is strongly enhanced in comparison with the results of Sec. VI A. This is a consequence of the slow dark-state polaritons, which require that each probe photon during its lifetime excites on average multiple atoms. As such, while the field content of the two contributions $\Sigma_{E}^{R1}$ and $\Sigma_{E}^{R2}$ as well as the relative detunings between atoms, lasers and guided photons, allow no distinction between these contributions, $\Sigma_{E}^{R2}$ is favored combinatorically by a factor $\sim C_P$. Consequently, slow polaritons with
FIG. 20. Flow diagram of the effective relative coupling strength $|1 + \chi|$ as a function of the externally adjustable parameter $\Omega_s$, where apart from $\gamma_d = 10$ the same parameters as in Fig. 12 are used together with the lenient interpretation of the non-linear Feynman rules. Note the emergence of a tristable region, where in addition to the opaque and transparent phases a new, strongly interacting semi-transparent phase appears.

FIG. 21. Same diagram as in Fig. 20 but using the strict version of the non-linear Feynman rules. Infinitely ranged interactions are typically dominated by the last diagram in the third line of Fig. 19.

As is indicated by the color gradients in Fig. 22, the transparent and opaque phase are adiabatically connected. The same is true for the transparent and intermediate phase as the latter emerges from the former at large drive strengths $\kappa_s$. In order to more closely investigate the properties of each phase, we provide a plot of the number density of atoms in the state $|s\rangle$ (Fig. 23), which shows that in every phase the polariton density and therefore their lifetime decreases as $\Omega_s$ is increased. However, in case of the intermediate phase $n_s$ and the polariton lifetime decrease also with increasing $\kappa_s$, which implies that the interaction strength is increased. This demonstrates that the intermediate phase is indeed stabilized by the overcompensation of $\Omega_s$ via strong interactions and its properties are not directly linked to either the weakly interacting limit $\Omega_s/\kappa_s \to \infty$ or the unperturbed polaritons at $\kappa_s/\Omega_s \to \infty$. We therefore use the strong backaction condition of a negative slope in the
polariton density $d\omega_s/d\kappa_s$ as the defining property to distinguish between the transparent and intermediate phase in Fig. 23. The relatively low density and the increased line-width of the dark-state polaritons (see Fig. 24) in the intermediate phase actually helps with the numerical investigation, as the discretization of momentum and frequency space can be performed at a lower resolution and saturation effects can more readily be discarded.

FIG. 22. The quantitative phase diagram in the limit $L_E \to \infty$ and with the parameters of Fig. 20, shows three distinct phases. While the transparent and opaque phase can be adiabatically connected to free theories far away from the multistable regime, the same cannot be said for the strongly interacting intermediate phase. The region of coexistence between opaque and transparent phase is indicated in magenta, that between transparent and intermediate phase in orange, and the remaining bistable area in green. All multistable regions are labeled by the initial characters of the coexisting phases.
FIG. 23. The number density of atoms in the metastable state $|s\rangle$ can be used to characterize the three distinct phases. For the slow polaritons obtained for the parameters of Fig. 20 that are also used here, $n_s$ is a good estimate of the dark-state polariton density. The density of the intermediate phase is highlighted in green and those of the adiabatically connected transparent and opaque phases in blue. If these coexist the transparent solution is shown in yellow. As a testament to the overcompensation of $\Omega_s$ by $\chi$ the density of the strong coupling intermediate phase decreases as drive intensity $\kappa_s$ is increased.

VII. CONTROLLED EXPANSION TO FINITE $L_E$

As was summarized at the end of the last section, the restriction to a theory that exclusively resums all Hartree diagrams of the effective theory in Fig. 19 is not always quantitatively justified. In particular, for current experiments with PCWs the range of the exchange photons is limited due to imperfections in the fabrication that cause rather large losses $\kappa_E$. Therefore, in this section we will go one step further and include all diagrams in next-to-leading order. This allows us to include scattering between polaritons, that is,
FIG. 24. Comparison of the EIT window for the two different stable phases shows a distinct ordering in the brightness of the dark-state polaritons. Except for $\kappa_s = 2$ and $\Omega_s = 0.21$ the parameters of Fig. 20 were used.

processes involving momentum transfer. In terms of the effective theory in Fig. 19 the only modification is the inclusion of the two diagrams in Fig. 25. Equivalently, in terms of the original theory including the atomic degrees of freedom, we obtain the Dyson equations shown in Fig. 26. One can identify these self-energies with the full set of self-consistently generated diagrams from the next-to-leading order corrections in $1/L_E$ and $1/L_P$ to the probe-photon propagator shown in Fig. 9.

FIG. 25. Addition to the effective Theory in Fig. 19 at next-to-leading order in $1/L_E$.

**A. Self-consistent theory at $O(1/L_E)$**

As can be seen in Fig. 26 a fully self-consistent theory involving all effects at next-to-leading order in the inverse interaction range requires to solve an even larger number of
FIG. 26. Complete set of coupled Dyson equations at next-to-leading order. The loop-reduction procedure of section III E is employed here and, depending on the Feynman rules at use, $d$-propagators are either bare or given as part of $G_{sd}$, of which the $s$-propagator in the last diagram of the third line is just the $(11)$-component.

This task might seem daunting at first sight, however, using the Kramers-Kronig relations (see section III C) and the loop reduction procedure (see section III E), every single diagram can once again be broken down into a combination of independent one-loop effects. Due to the non-linear Feynman rules
FIG. 27. Loop reduction procedure for the Feynman diagram in Fig. 9b), which formally delocalizes excited atoms. Note that during this procedure the probe photon coupling strength has to be modified by $g_P \to g_P \sqrt{1 - 2n_V}$ to properly reflect the atom number density.

(section III D), great care has to be taken in determining which of these single loop effects can be combined. We do so by introducing two different matrix Green’s functions $G_{sd}$ and $\tilde{G}_{sd}$ for the states $|s\rangle$ and $|d\rangle$. To help distinguish these propagators in Feynman diagrams, we slash the propagator of $\tilde{G}_{sd}$ twice. When appearing as an insertion inside the probe-photon propagator, $G_{sd}$ cannot itself involve a self-energy that would return the atom to its ground state. There is thus only one contribution to the self-energy and the matrix propagator takes the fairly simple form

$$G_{sd}^{R/K} = \left( [G_{sd}^{R}]^{-1} - \Sigma_{sd}^{R} \right)^{-1},$$

(95)

where

$$\Sigma_{sd}^{R} = \frac{i}{2} g E \left( \delta G_{E_{22}}^{K} \ast \tilde{G}_{dd}^{R} + G_{E_{22}}^{R} \ast \delta \tilde{G}_{dd}^{K} + G_{E_{21}}^{R} \ast \delta \tilde{G}_{ds}^{K} + \left( \delta G_{E_{21}}^{K} + 2G_{E_{21}}^{K0} \right) \ast \tilde{G}_{ds}^{R} \right),$$

(96)

uses $\ast$ to denote the convolution in $\omega$ and $k$. The corresponding Keldysh component reads

$$\delta G_{sd}^{K} = G_{sd}^{R} \cdot \delta \Sigma_{sd}^{K} \cdot G_{sd}^{A},$$

(97)

with

$$\Sigma_{sd}^{K} = \frac{i}{2} g E \left( \delta G_{E_{22}}^{K} \ast \delta \tilde{G}_{dd}^{K} \ast \delta \tilde{G}_{ds}^{K} \right).$$

(98)

Note that $\Sigma_{sd}^{R/K}$ is allowed to depend on the quasi-momentum $k$, since, due to the photon admixture with momentum transfer, it effectively no longer describes a completely stationary atom. This effective delocalization of the atoms is not an actual physical process, but rather a mathematical trick to accommodate the loop reduction procedure shown in Fig. 27. In
\[ \tilde{G}_{sd} = \left( [\tilde{G}^R_{sd}]^{-1} - \begin{pmatrix} \Sigma^R_s & 0 \\ 0 & 0 \end{pmatrix} \right)^{-1} \]  

(99)

with

\[ \Sigma^R_s = \frac{\Omega^2}{\omega - g_P^2 \left[ (1 - n_V)G^R_P + \frac{1}{2} G^R_s \ast \omega \delta G^K_P \right] + i \gamma_c / 2}, \]

(100)

where \( \ast \omega \) indicates a particle-hole convolution in frequency only, i.e.

\[ f \ast \omega h = \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} f(\omega')h(\omega' + \omega). \]

(101)

Furthermore, the related

\[ \tilde{G}_{sd}^K = \tilde{G}_{sd}^R \cdot \begin{pmatrix} \delta \Sigma^K_{sd} & 0 \\ 0 & 0 \end{pmatrix} \cdot \tilde{G}_{sd}^R, \]

(102)

where

\[ \delta \Sigma^K_s = \frac{g_P^2}{\Omega^2} (1 - n_V)\delta G^K_P |\Sigma^R_s|^2 \]

(103)

correctly includes all repeated scattering processes of a probe photon into a probe and an exchange photon. The loop reduction procedure therefore allows for a very cost-efficient inclusion of pairing effects between probe and exchange photons, that would otherwise require a self-consistent treatment of the corresponding T-matrix.

The Dyson equation for the probe photon propagator takes almost exactly the same form as it does for the non-interacting EIT:

\[ G^R_P = \left( \omega - \Delta_P(k) - \frac{g_P^2 (1 - n_V)}{\omega - \Omega^2 G^R_{ks} + i \gamma_c / 2} + i K_P / 2 \right)^{-1} \]

\[ G^K_P = \left( \Omega^2 \delta G^K_{ss} | G^R_e |^2 - 2i \kappa_s(\omega) \right) | G^R_P |^2, \]

(104)

with the only difference being hidden in the more elaborate form of \( G_{ss} \).

In order to close the set of coupled equations one has to find the full exchange photon propagator, which again has two self-energy contributions as in Eq. 92. The first one

\[ \Sigma^R_E(\omega) = \frac{i}{2} g^2_E \begin{pmatrix} \delta G^K_\delta \ast \omega G^R_d(\omega) & 0 \\ 0 & (\delta G^K_\delta \ast \omega G^R_d)(-\omega) \end{pmatrix} \]

(105)
is already known from Sec. VI A and in the present notation involves the propagator $\delta G^K_\delta$ given by

$$
\delta G^K_\delta(\omega) = \frac{g_p^2}{\Omega^2} (1 - n_V) \kappa_s(\omega) \left| \frac{\Sigma^R(\omega)}{\omega - \Delta_s - \Sigma^R(\omega) + i\gamma_s/2} \right|^2 \int \frac{dk}{2\pi} \delta G^K_\delta(\omega, k).
$$

(106)

For the strict interpretation of the non-linear Feynman rules we use

$$
\Sigma^R(\omega) = \frac{\Omega^2}{\omega + i\gamma_s/2 - g_p^2 \int \frac{dk}{2\pi} [(1 - n_V)G^R_\delta + \kappa_s(\omega)G^R_\delta * \delta G^K_\delta]}
$$

(107)

and $G^R_\delta = G^R_{d_0}$. One therefore recovers exactly the same expression for $\Sigma^R_{E_1}$ as in the previous sections. In case of the lenient Feynman rules the denominator in the absolute value in Eq. (106) is to be replaced by

$$
\omega - \Delta_s - \Sigma^R(\omega) + i\gamma_s/2 + \frac{(\Omega_s^\text{eff})^2}{\omega - \Delta_d - \Delta_s + i\gamma_d/2} + \int_{-\pi}^\pi \frac{dk}{2\pi} \Sigma^{R_s}(\omega, k),
$$

(108)

where $\Sigma^{R_s}$ is the 11-component of $\Sigma^R_{sd}$. At the same time $G^R_\delta$ is given by $\tilde{G}^R_{dd}$. This leaves $\Sigma^R_{E_2}$, which takes the same form as in Sec. VI B 2.

$$
\Sigma^R_{E_2}(\omega, k) = \frac{i}{2} g_p^4 g_E^2 \Omega^4 (\Omega_s^\text{eff})^2 (1 - n_V)^2 \int \frac{d\omega' dp}{2\pi 2\pi} |G^R_e(\omega')G^R_s(\omega')|^2 \delta G^K_\delta(\omega', p) \times
$$

$$
\begin{bmatrix}
[G^R_e(\omega + \omega')G^R_e(\omega + \omega')]^2 |G^R_d(\omega + \omega')|^2 & G^A_d(\omega')G^R_d(\omega + \omega') \\
G^R_d(\omega')G^R_d(\omega + \omega') & |G^R_d(\omega')|^2
\end{bmatrix}
$$

$$
+ [G^A_e(\omega' - \omega)G^A_s(\omega' - \omega)]^2 |G^R_d(\omega')|^2 \times
$$

$$
\begin{bmatrix}
|G^R_d(\omega')|^2 & G^A_d(\omega' - \omega)G^A_d(\omega') \\
G^A_d(\omega' - \omega)G^A_d(\omega') & |G^A_d(\omega' - \omega)|^2
\end{bmatrix}
$$

(109)

Evaluating the same diagrams for the Keldysh component, one obtains the last two pieces of the puzzle:

$$
\Sigma^R_{E_1} = 0,
$$

(110)

as before, and

$$
\Sigma^R_{E_2}(\omega, k) = \int \frac{d\omega' dp}{2\pi 2\pi} |G^R_e(\omega')G^R_e(\omega' + \omega)G^R_s(\omega' + \omega')|^2 \
\times \delta G^K_\delta(\omega', p) \delta G^K_\delta(\omega' + \omega, p + k) \begin{bmatrix}
|G^R_d(\omega' + \omega)| & G^A_d(\omega')G^A_d(\omega' + \omega') \\
G^A_d(\omega' + \omega') & |G^A_d(\omega' + \omega')|^2
\end{bmatrix}
$$

(111)
with the same choices for $G^R_d$ in $\Sigma_{E}^{R/K}$ as in $\Sigma_{E}^{R_1}$.

As always, throughout this entire theory the bare laser coupling $\Omega_1$ between states $|s\rangle$ and $|d\rangle$ has been replaced by $\Omega_{s}^{\text{eff}} = \Omega_1|1 + \chi|$, where $\chi$ given by (94) describes the modified conversion rate between $|s\rangle$ and $|d\rangle$ due to the presence of other polaritons, as in section VI A. Con-}

FIG. 28. Dependency structure and ordering of updates for the self-consistent solution of the
Dyson equations (95) through (111) with (94) iteratively updating the effective coupling $\Omega_{s}^{\text{eff}}$.

trary to the previous renditions of the self-consistent structure, with the inclusion of $1/L_E$ effects, scattering of probe photons into exchange photons becomes a possibility. Therefore, self-consistence is no longer simply a question of finding the right parameter $\chi$, but actually involves the full frequency and momentum dependent Green’s functions $G^R_E$. As such, the numerical implementation has to find the solution in an iterative manner. Since there is no direct dependence on $\Omega_1$, one can however fix $\Omega_{s}^{\text{eff}}$, initialize all Green’s functions as bare ones and iterate equations (95) through (111) together with (94) until $\chi$ no longer changes, see Fig. 28. This once again means that the final value of $\Omega_s$ corresponding to the solution is not known a priori and has to be searched for iteratively, unless the entire phase diagram is calculated. The main advantage of this method lies again in the enhanced convergence that is unaffected by the presence of any phase transition.

As we have already discussed in Sec. VIB2, the use of the absolute value in the definition of $\Omega_{s}^{\text{eff}}$ in the anomalous Green’s functions is wrong, since $G_E$ no longer transforms correctly under a global $U(1)$ gauge transformation. Previously this was not a problem for the evaluation of gauge invariant observables. Despite the iterative procedure the backaction at any stage of the iteration for any observable depends only on $|1 + \chi|^2$ as is required by gauge
invariance. The described self-consistent calculation thus finds the correct value of $|1 + \chi|$ and therefore of all normal GFs. To also obtain the anomalous components the correct phase of $\chi$ simply has to be restored in the final result.

B. Results

When including the effects of a finite interaction range, care has to be taken as not to break any of the assumptions underlying the quantitative validity of the approximations at use. In particular, if the interaction becomes too short-ranged, the losses in state $|d\rangle$ caused by emission of exchange photons and described by the second diagram of the fourth and fifth line of Fig. 26 – or equivalently the (22)-component of Eq. 96 – become large as a result of the narrow line-width of state $|s\rangle$ for long lived dark-state polaritons. These effects are included in $G^R_d$ in the lenient interpretation of the non-linear Feynman rules, but not for the strict rules. As these atomic Green’s functions form the vertex of the effective theory, the differences will grow upon iteration of the self-consistency equations. The uncertainty regarding the results of the exact Feynman rules for four-level atoms thus grows with decreasing $L_E$. This is already observable in the comparison between Fig. 29 and Fig. 30 which qualitatively show the same phases, but with a larger discrepancy in the actual phase boundary than in the previous sections. Since the additional scattering effects that arise from the inclusion of $1/L_E$ effects into the description cannot themselves create any new instabilities and instead remedy those that could otherwise exist in $G^R_E$, relatively large values of $C_E$ can be treated without much more than quantitative corrections to the previously discussed results. In particular, the parameters discussed in Figs. 29 and 30 correspond to $C_E \approx 0.22$ and $C_P = 2$. The main limitation for an extension to even smaller values of $L_E$ or larger values of $C_E$ lies in the discrepancy between the different interpretations of the Feynman rules, which eventually will have to be specified in more detail.

This time, for a change, we discuss our results using numerical data obtained from the lenient Feynman rules, which requires exactly the same amount of numerical effort as the strict rules. The resulting phase diagram depicted in Fig. 31 is very similar to that in Fig. 22. However, the quantitative corrections due to the finite interaction range result in a small area where all three phases are stable.
The restriction to large interaction ranges imposed by the discrepancy between the

FIG. 29. Flow diagram of the effective relative coupling strength $|1 + \chi|$ as a function of the externally adjustable parameter $\Omega_s$, using the same parameters as in Fig. 20 except for $\alpha_E = 400$ and $k_E = 0$, that previously did not need to be specified. Here we use the lenient interpretation of the non-linear Feynman rules. Note that the qualitative structure remains the same as in Figs. 20 and 21 however the quantitative differences compared to the strict rule in Fig. 30 has increased.

FIG. 30. Same diagram as in Fig. 29 but using the strict version of the non-linear Feynman rules.

approximate implementations of the Feynman rules, together with the fact that scattering between dark-state polaritons is dominated by forward scattering – the exchange photons are most efficiently coupled to at $k = 0$ – renders the effects of scattering on the probe photons actually negligible in this regime. As a demonstration of the smallness of the redistribution due to scattering, one can examine the distribution function $F_P(\omega, k)$, which, even in the transparent phase where resonant scattering is strongest, is almost entirely momentum independent (see Fig. 32). Only upon subtraction of the momentum independent background a slight increase in $F_P(\omega, k)$ near the EIT window can be observed. As such, there is also no significant deformation in the dispersion of the dark-state polariton (Fig. 33) and the the number-density of dark-state polaritons experiences only minor corrections (Fig. 34).
FIG. 31. Phase diagram including corrections due to the finite interaction range. The color coding is the same as in Fig. 22. The bistability between transparent and intermediate phase is more pronounced, giving rise to a tristable regime colored in violet. The parameters are identical to those in Fig. 22 except for $\alpha_E = 1000$ and $k_E = 0$ (setting the interaction range and profile) and the use of lenient Feynman rules.

For the present case of scattering with small momentum transfers, the most significant effect of the inclusion of $1/L_E$ corrections is the avoidance of the divergence in $G_E^R$ appearing as an artifact of the $L_E \to \infty$ theory: while the exchange photons can still experience an effective drive due to the redistribution of energy between dark-state polaritons, this effect
is significantly weakened by the increasing dissipative nature of the atomic vertex brought about by the aforementioned losses in $|d\rangle$. As the exchange photon experiences fewer and fewer losses, those of $|d\rangle$ namely increase, thereby weakening the coupling between probe and exchange photons enough to stabilize the system.

![Graph](image)

FIG. 32. For parameters where the expansion remains quantitatively controlled, the distribution function $F_P(\omega, k)$ shows hardly any visible momentum dependence and thus only weak signatures of scattering. To make the weak momentum dependence visible, we subtracted the momentum independent background $F_P(\omega, k = 0)$. Here the transparent solution is depicted for the same parameters as in Fig. 29 with $\kappa_s = 1.44$ and $\Omega_s = 0.15$.

![Graph](image)

FIG. 33. Using the same parameters as in Fig. 29 except for $\alpha_E = 1000$, $k_E = 0$, $\kappa_s = 1.44$ and $\Omega_s = 0.15$, one again notes the pronounced difference in the overall density between the three stable phases. In particular, the opaque phase contains orders of magnitude fewer polaritons than the other two.
FIG. 34. The photon density near the EIT condition is proportional to the atom density in the metastable state \( n_s \) and changes only insignificantly relative to the results for \( L_E \to \infty \) if the same parameters are used (here those of Fig. 29).

VIII. COMPARISON WITH POLARITONS IN RYDBERG ENSEMBLES

Rydberg atoms exhibit essentially the same level-structure as the atoms we previously considered, but without the excited state \( |d\rangle \). We will therefore mostly use the same notation as above to illustrate how interactions between Rydberg polaritons fit into a \( 1/L \) expansion. Instead of exchange photons with significant losses and a tunable dispersion, zero point fluctuations in the electromagnetic field mediate the interactions between Rydberg atoms. In fact the quadratic Stark shift that gives rise to the interatomic van-der-Waals potential \( V(x) = C/|x|^6 \) requires the exchange of two photons. Their dynamics however happens on timescales much shorter than those experimentally relevant and can therefore be neglected. With this knowledge it is well justified to replace the two-photon interaction by the effective potential \( V(x - x')n_s(x)n_s(x') \).

Diagrammatically, the resulting theory looks very similar to the one discussed in the previous sections, the only modification being the replacement of \( G_R^E \) coupling between states
FIG. 35. Leading order of the expansion in Feynman diagrams around the limit of infinitely ranged interactions between Rydberg polaritons. Note that, due to the use of a fixed potential, only a single interaction diagram has to be considered. Otherwise the self-consistent treatment is similar to that in Sec. VI A.

|s⟩ and |d⟩ by V(x) acting directly on |s⟩. The non-interacting Rydberg polariton theory is illustrated in the first line of Fig. 35 if the |s⟩-propagator is considered as bare. Interactions are then taken into account by dressing this state with density-density interactions, that take a similar form as those considered in Sec. VI A. The resulting Feynman diagram in the second line of Fig. 35 has been treated self-consistently following the same procedure as in Sec. VI A, with the main difference compared to Fig. 10 being the absence of state |d⟩ and the external source Ωs. It is readily evaluated as

$$\Sigma_s^R(x, t) = \int d^dx V(x - x') n_s(x', t)$$

with the functional dependencies $G_s^R [\Sigma_s^R]$, $G_e^R [G_s^R]$ and $G_P^R [G_e^R]$ identical to Sec. VI A. Interestingly, the leading diagrammatic contributions for the setups discussed in the previous sections actually disappear in the context of Rydberg polaritons. Self-interactions of a Rydberg atom by emission and absorption of a zero-point fluctuation induce a Lamb shift.
that is already included in the bare energy of the atomic state. A repeated interaction between two Rydberg atoms on the other hand has to be treated with the non-linear Feynman rules. By arguments identical in spirit to those of section III D it reduces to terms already included in (112). Last but not least, a self-interaction of a Rydberg polariton through the interaction of two distinct atoms, similar to Fig. 9b), is excluded by the instantaneous nature of interactions.

As such, the low density limit of Rydberg polaritons results in a less complicated, but conceptually similar expansion to that derived for PCWs and TNWs. However, the absence of an external coupling similar to $\Omega_s$, with which the interaction can destructively interfere, prevents the emergence of phase transitions of the type discussed before.

Instead, interesting questions include the scattering of Rydberg polaritons and the stability of regular structures (i.e. n-particle bound states) or even crystals. Here we only want to give a brief idea of how these questions can be approached in terms of a $1/L$ expansion and therefore discuss the simple case of a Rydberg polariton scattering off a fixed Rydberg atom at the origin. In this case the polariton Green’s function is given by

$$G_R^P(\omega) = (\omega - \omega_P(k) - \frac{g_P^2(1 - n_V)}{\omega - \frac{\Omega^2}{\omega - \Delta_s - V(|x|)} + i\gamma_e/2} + i\kappa_M/2)^{-1}, \quad (113)$$

where the inversion in momentum space is the inverse with respect to the convolution and thus a non-trivial operation. Nevertheless assuming a slow, and thus well-localized, incoming Rydberg polariton of fixed frequency $\omega_{EIT} = \Delta_s$ corresponding to the EIT window at $|x| \rightarrow \infty$, we can calculate its losses as a function of $r = |x|$ and determining the blockade radius. The losses are given by the imaginary part of the inverse propagator

$$\kappa_{\text{eff}}(\omega, r) = \kappa_P + \frac{g_P^2(1 - n_V)\gamma_e}{\omega - \frac{\Omega^2}{\omega - \Delta_s - V(|x|)} + \gamma_e^2/4}, \quad (114)$$

and illustrated in Fig. 36. The pronounced maximum that forms at the blockade radius $R_b$ is determined by equating the frequency shift due to $\Sigma^R_e$ with the bare losses $\gamma_e/2$. As a result one finds

$$R_b = \left(\frac{C(\gamma_e + 2\Delta_s)}{2\Omega^2}\right)^{1/6} \quad (115)$$

Finally, an expansion of $\kappa_{\text{eff}}(\Delta_s, r)$ around $r \rightarrow \infty$ reproduces the well-known result $\kappa_P^\text{eff}(\Delta_s, r) \propto r^{-12}$. 

66
FIG. 36. Rydberg blockade experienced by a Rydberg polariton as a function of the distance from a stationary Rydberg atom at $r = 0$. The imaginary part of the inverse propagator indicating the losses becomes very large at a distance set by the blockade radius $R_b$. At the same time the real part that gives rise to deflection also grows. Parameters used are $\gamma_e = 1/4$, $\kappa_P = 1/5$, $\Delta_s = 1/3$, $\Omega = 1/2$, $n_V = 0$ and $g_P = 5$.

IX. CONCLUSIONS AND OUTLOOK

We have developed a non-equilibrium diagrammatic approach to strongly interacting photons in optically dense atomic media. It provides quantitative results in the regime of low atom-excitation densities where saturation effects are negligible, provided the atom-atom interactions – which the photons inherit – have a large effective range compared to the atomic interspacing. Such a regime can still feature single-atom cooperativities of order or larger than one and thus show strong nonlinearities and emergent many-body phenomena, like the non-equilibrium phase transitions we describe here.

The formalism applies to a broad class of hybrid (nano)photonic devices coupled to arrays of interacting atoms and constitutes a novel theoretical approach to such driven-dissipative many-body systems. Important future applications involve in first place the description of experiments where photon wavepackets propagate through the medium, the description of
which requires to solve the Dyson’s equations presented above without relying on space- and
time-translation invariance. A second interesting avenue is the study of possible crystalline
phases of photons appearing in the steady-state\cite{10,11}, which can be also efficiently described
within our formalism.

X. ACKNOWLEDGMENTS

DC acknowledges support from ERC Starting Grant FOQAL, MINECO Plan Na-
cional Grant CANS, MINECO Severo Ochoa Grant No. SEV-2015-0522, CERCA Pro-
gramme/Generalitat de Catalunya, and Fundacio Privada Cellex.

XI. SUPPLEMENTAL MATERIAL

A. Alternative derivation of non-linear Feynman rules

In section III D we argued that in order to capture the properties of non-interacting po-
laritons giving rise to electromagnetically induced transparency, it is not actually necessary
to implement non-linear Feynman rules in real time. Instead, it suffices to simply exclude
all self-energy insertions into the ground-state propagator and to forbid any of the excited
states to repeatedly couple to the ground-state. By these means we then derived the (exact)
polarizability of the atomic medium in IV. It is instructive to rederive this result directly
from the coupled Lindblad equations of the spin operators $\sigma_{\mu,\nu}$ introduced in Sec. III A.
The retarded polarizability of each atom in the ground-state is given by

$$P(t) = \theta(t) \left[ \text{Tr}(\sigma_{g,e}(t)\sigma_{e,g}\rho) - \text{Tr}(\sigma_{e,g}\sigma_{g,e}(t)\rho) \right].$$

(116)

The latter of these contributions vanishes identically as $\rho = \sigma_{g,g}$. The time-evolution of
$\sigma_{g,e}(t)$ is given by

$$-i\dot{\sigma}_{g,e}(t) = [H, \sigma_{g,e}(t)] + i\gamma_e \left( \sigma_{g,e}\sigma_{g,e}(t)\sigma_{e,g} - \frac{1}{2}\{\sigma_{e,g}\sigma_{g,e},\sigma_{g,e}(t)\} \right),$$

(117)

which very nicely simplifies, if one uses $\sigma_{e,g}\sigma_{g,e} = \sigma_{e,e}$ as well as the observation that $H$ in the
limit of low polariton densities acts trivially on the ground-state, which implies $\sigma_{g,e}\sigma_{g,e}(t) = 0$ and
$H\sigma_{g,e}(t) = 0$. One thus ends up with

$$i\dot{\sigma}_{g,e}(t) = \sigma_{g,e}(t)H - i\frac{\gamma_e}{2}\sigma_{g,e}(t)\sigma_{e,e},$$

(118)
which has the solution

\[ \sigma_{g,e}(t) = \sigma_{g,e} e^{-i\tilde{H}t}, \]  

where \( \tilde{H} = H + i\frac{\gamma_e}{2} \sigma_{e,e} \) is the non-hermitian effective Hamiltonian governing the time-evolution of the three level system in the presence of losses. Inserting this result back into the polarizability, we obtain

\[ P(t) = \theta(t) \left( e^{i\tilde{H}t} \right)_{22}, \]  

which upon Fourier transformation turns into

\[ P(\omega) = \frac{i}{\omega - \Omega^2 + i\gamma_d/2}, \]  

which coincides with the result obtained by means of the simplified non-linear Feynman rules in [46]. We thus have seen that, due to the absence of laser coupling between the atomic ground-state and the excited state, non-linear Feynman rules are easily implemented. As is mentioned in Sec. III D, the dynamics of the other external state of the N-scheme is not as simple. Thus a similar derivation for the susceptibility of the medium from the perspective of the exchange photon fails.

We can now go beyond the limit of low polariton densities and consider the effect of a finite density of excited atoms. In this case, there is a finite fraction of atoms for which \( \rho \neq \sigma_{g,g} \).

In particular due to EIT, excited atoms will predominantly occupy the metastable state \( (\rho = \sigma_{s,s}) \). This however implies that \( \sigma_{e,g}\rho \) and \( \rho\sigma_{e,g} \) both vanish. The main effect of a finite density of polaritons will thus simply result in a reduced susceptibility of the atoms. This is easily included via a finite density of defects \( n_V \) in the chain of atoms. If the number density of excited atoms remains small, this effect can be neglected all together, as it will have no effect on the stability of phases reported here.

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