Inelastic photon scattering via the intracavity Rydberg blockade

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Electromagnetically induced transparency (EIT) in a ladder system involving a Rydberg level is known to yield giant optical nonlinearities for the probe field, even in the few-photon regime. This enhancement is due to the strong dipole-dipole interactions between Rydberg atoms and the resulting excitation blockade phenomenon. In order to study such highly correlated media, ad hoc models or low-excitation assumptions are generally used to tackle their dynamical response to optical fields. Here, we study the behaviour of a cavity Rydberg-EIT setup in the non-equilibrium quantum field formalism, and we obtain analytic expressions for elastic and inelastic components of the cavity transmission spectrum, valid up to higher excitation numbers than previously achieved. This allows us to identify and interpret a polaritonic resonance structure, to our knowledge unreported so far.

PACS numbers: 42.50.Ar, 32.80.Ee, 42.50.Gy, 42.50.Nn

Introduction. Optical quantum information processing requires photonic gates. For the sake of efficiency it is preferable to implement them in a deterministic way, which implies photon-photon interactions. Though impossible to achieve directly, such interactions can be effectively emulated by coupling photons to an atomic ensemble driven in a Rydberg-EIT configuration either in free-space 1,2 or in a cavity setup 3, 4. The full dynamics of such a strongly correlated many-body system and therefore its effects on the incoming photons cannot be computed exactly. So far, analytic expressions of dynamical variables like, e.g., the correlation functions of the transmitted field could be derived either using ad hoc models – such as the Rydberg bubble picture 1, 2, or resorting to the perturbation theory restricted to the lowest non-vanishing order in the number of incoming photons 3, 4. In this Letter, we employ the Schwinger-Keldysh contour formalism 7, 8 to derive analytic expressions for field correlation functions in a cavity setup beyond the lowest non-vanishing order in the excitation number 6. By opening a systematic and manageable way to deal with higher-order terms, our approach breaks new ground for solving the outstanding problem set by the many-body dynamics of Rydberg-blockaded ensembles interacting with quantized light. It also allows us to unveil nontrivial physical features of the transmitted light spectrum that we explain by a simple polaritonic picture.

We consider an ensemble of $N$ atoms with a ground, intermediate and Rydberg states, denoted by $|g\rangle$, $|e\rangle$ and $|r\rangle$, respectively, loaded in an optical cavity 5. The transitions $g \leftrightarrow e$ and $e \leftrightarrow r$ are driven by the cavity mode, of frequency $\omega_{c}$ and annihilation operator $a$, and the strong control field, with the coupling strength $g$ and the Rabi frequency $\Omega_{cf}$, respectively. The cavity is fed through an input mirror with decay rate $\gamma_{c}^{(d)}$; we moreover set $\gamma_{c} = \gamma_{c}^{(f)} + \gamma_{c}^{(d)}$. We define detunings for the cavity $\Delta_{c} = (\omega_{p} - \omega_{c})$, single-photon $\Delta_{1} = (\omega_{p} - \omega_{eg})$ and two-photon $\Delta_{2} = (\omega_{p} + \omega_{cf} - \omega_{rg})$, with respect to the frequencies $\omega_{eg}$ and $\omega_{rg}$ of the $g \leftrightarrow e$ and $g \leftrightarrow r$ transitions. We denote by $\gamma_{c}$ and $\gamma_{r}$ the decay rates from the intermediate $|e\rangle$ and Rydberg $|r\rangle$ states, respectively.

If there were no atomic interactions, the cloud driven under perfect EIT conditions ($\gamma_{c} \approx \Delta_{c} \approx 0$), would be transparent for the probe light. The dipole-dipole-interaction-induced blockade phenomenon 10, 11 actually prevents most of the atoms in the sample from being Rydberg excited. If $\Delta_{c} \approx 0$, spontaneous emission from the intermediate state is strongly enhanced which significantly modifies the shape of the transmitted light spectrum. This effect can be characterized by the steady state correlation function of the intracavity light $\langle a^\dagger(t) a(0) \rangle$ 12. At the lowest non-vanishing order in the feeding rate $|a| \equiv \sqrt{2\gamma_{c}^{(f)}} I_{in}$, where $I_{in}$ is the incident photon flux fed into the cavity, the correlation function was shown to factorize, i.e. $\langle a^\dagger(t) a(0) \rangle = \langle a^\dagger(t) \rangle \langle a(0) \rangle$, where the superscript denotes the order in $a$. To reveal nonlinear features, one has to investigate orders higher than four – by conservation of excitation number the third order vanishes. Usual techniques are not suited to this task. In particular, the standard fourth-order perturbative expansion would already lead to a cumbersome hierarchy of Heisenberg equations which could hardly be generalized further. Here, we show the Schwinger-Keldysh contour formalism 7, 8 allows one to compute dynamical variables of the system up to a priori arbitrary order in the feeding strength, in a systematic and handy way. Besides bringing physical insight into the specific problem considered here, our calculation demonstrates how powerful this approach is to deal with non-equilibrium dynamics of atomic systems as already stressed in 13.
Dynamical equations of the system. According to Holstein-Primakov approximation, the atomic lowering operators \( \sigma_{j\alpha}^{(n)} \) and \( \sigma_{j\alpha}^{(n)} \) can be treated as bosons \( b_n \) and \( c_n \), respectively, in the low excitation regime [34]. Performing the rotating wave and Markov approximations, the relevant Heisenberg-Langevin equations are

\[
\frac{d}{dt}a = i[H, a] - \gamma_a a + \sqrt{2\gamma_c(f)} a_{in}^{(f)} + \sqrt{2\gamma_c(d)} a_{in}^{(d)} \tag{1}
\]

\[
\frac{d}{dt}b_n = i[H, b_n] - \gamma_c b_n + b_{in,n} \tag{2}
\]

\[
\frac{d}{dt}c_n = i[H, c_n] - \gamma_c c_n + c_{in,n} \tag{3}
\]

where \( \{a_{in}^{(f)}, a_{in}^{(d)}, b_{in,n}, c_{in,n}\} \) denote the respective Langevin forces associated to the incoming fields from the feeding and detection sides, and to the atomic operators \( b_n \) and \( c_n \). The Hamiltonian of the system \( H = H_0 + H_{int} \) comprises the linearized EIT (later referred to as “unperturbed”) Hamiltonian

\[
H_0 = -\Delta_c a^\dagger a - \sum_{n=1}^{N} (\Delta_c b_n^\dagger b_n + \Delta_c c_n^\dagger c_n) + \sum_{n=1}^{N} \left( g a^\dagger b_n + \frac{\Omega_f}{2} c_n^\dagger b_n + H.c. \right) \tag{4}
\]

and the so-called interaction Hamiltonian \( H_{int} = H_{dd} + H_f \) consisting of the dipole-dipole interaction Hamiltonian \( H_{dd} = \sum_{m<n}^{N} \kappa_{mm} c_n^\dagger c_m c_m^\dagger c_n \) and the cavity feeding Hamiltonian \( H_f = \alpha (a + a^\dagger) \), where \( \kappa_{mm} \equiv C_6/\|\vec{r}_m - \vec{r}_n\|^6 \). In the following, we will need the operators expressed in the interaction picture, i.e. the solutions \( \{a_0(t), b_{n0}(t), c_{n0}(t)\} \) of the system Eqs. (1) in which \( H \) is replaced by \( H_0 \).

Correlation functions in the Schwinger-Keldysh contour formalism. The correlation function \( \langle a^\dagger(t_1) a(t_2) \rangle \), computed in the vacuum state \( \rho_0 = |0\rangle \langle 0| \), can be put in the form

\[
\text{Tr} \left\{ \rho_0 \mathcal{T} c \left( e^{-i[(\mathcal{J} H_{int})^t]_0} (t_1^\dagger) a_0 (t_2^\dagger) \right) \right\} \tag{5}
\]

where \( \mathcal{T} c \) is the ordering operator along the contour \( C^- \) comprising the forward \( C^+ \equiv (-\infty, \infty) \) and backward \( C^- \equiv (-\infty, -\infty) \) parts \( [3, 4] \). \( \mathcal{T} c \) is explicitly defined as follows, for generic Heisenberg operators of the system \( \{A_j(t)\} \),

\[
\mathcal{T} c \left( A_1 (t_1^\dagger) \cdots A_k (t_k^\dagger) A_{k+1} (t_{k+1}^\dagger) \cdots A_{k+l} (t_{k+l}^\dagger) \right) = \mathcal{T} \left( A_1 (t_1^\dagger) \cdots A_k (t_k^\dagger) \right) \times \mathcal{T} \left( A_{k+1} (t_{k+1}^\dagger) \cdots A_{k+l} (t_{k+l}^\dagger) \right)
\]

where \( \mathcal{T}, \mathcal{T} \) denote the usual chronological and antichronological ordering operators, respectively, and the superscript \( ^\dagger \) indicates the branch \( C^\pm \) the time argument belongs to. More generally, multitime correlation functions

\[
\left\langle \mathcal{T} \left\{ L_{n_1} \cdots L_{n_k} \right\} \right\rangle = \left\langle \mathcal{T} \left\{ L_{m_1} \cdots L_{m_l} \right\} \right\rangle
\]

involving operators \( L^j \) take the form

\[
\left\langle \mathcal{T} c \left( e^{-i[(\mathcal{J} H_{int})^t]_0} (t_1^\dagger) \cdots L_{n_k} (t_{n_k}^\dagger) \cdots L_{n_l} (t_{n_l}^\dagger) \right) \right\rangle
\]

where \( L_j^0 \) is the operator \( L^j \) expressed in the interaction picture. Fig. 1 represents the contour-ordering used for a specific correlation function.

Perturbative calculation of the correlation functions. We now perturbatively expand the correlation function Eq. (5) with respect to the feeding Hamiltonian, i.e. \( \langle a^\dagger(t_1) a(t_2) \rangle = \sum_{k=0}^{+\infty} \langle a^\dagger(t_1) a(t_2) \rangle^{(k)} \), where

\[
\langle a^\dagger(t_1) a(t_2) \rangle^{(k)} = \frac{(-i\alpha)^k}{k!} \times \left\langle \mathcal{T} c \left( \int (a_0 + a_0^\dagger)^k e^{-i[(\mathcal{J} H_{int})^t]_0} (t_1^\dagger) \cdots (t_{n_k}^\dagger) \right) \right\rangle
\]

and the superscript denotes the order \( k \) in the feeding rate \( \alpha \). In the expansion, only remain the terms which contain the same number of creation and annihilation operators, corresponding to even \( k \)'s. These terms can be further evaluated by perturbatively expanding them with respect to the dipole-dipole interaction Hamiltonian \( H_{dd} \). From now on, shall more specifically be interested in the temporal Fourier transform of the correlation function \( \langle a^\dagger(t_1) a(t_2) \rangle \). Following Wick’s theorem \([3, 8]\), the contribution of each order in \( H_{dd} \) is a sum of products of the Fourier transformed time-ordered non-perturbed Green’s functions defined by

\[
G_{x_0,y_0} [\omega] = -i \int dt e^{i\omega t} \left\langle \mathcal{T} (x_0(t) y_0^\dagger(0)) \right\rangle \tag{7}
\]

where \( (x_0, y_0) \) stand for any 2 bosonic operators of the system in the interaction picture. These functions may
be readily computed from the (linear) Bloch equations spanned by $H_0$.

The full resummation of the perturbation series in $H_{dd}$ can be performed for the first four orders in $\alpha$. For the second order, we get

$$\langle a^\dagger (\omega_1) a (\omega_2) \rangle^{(2)} = \langle a^\dagger (\omega_1) \rangle^{(1)} \langle a (\omega_2) \rangle^{(1)}$$  \hspace{1cm} (8)

where $\langle a (\omega) \rangle^{(1)} = \sqrt{2\pi}\alpha\delta (\omega) G_{a,a_0} [0]$ is the first order average value for the cavity mode annihilation operator. Note that Eq. (8) exhibits the factorization property we pointed out and used in a previous work \[6\]. By contrast, the fourth order $\langle a^\dagger (\omega_1) a (\omega_2) \rangle^{(4)}$ contains a factorized part

$$\langle a^\dagger (\omega_1) \rangle^{(1)} \langle a (\omega_2) \rangle^{(3)} + \langle a^\dagger (\omega_1) \rangle^{(3)} \langle a (\omega_2) \rangle^{(1)}$$  \hspace{1cm} (9)

but also an extra component, which shall be denoted by $\langle a^\dagger (\omega_1), a (\omega_2) \rangle^{(4)}$ – this term is actually a covariance and characterizes the interaction-induced non-classicality of the system. More explicitly, the calculations yield

$$\langle a (\omega) \rangle^{(3)} = \delta (\omega) \sqrt{2\pi}\alpha^3 \langle G_{s_0,a_0} [0] \rangle^2 T_0 \langle G_{a_0,a_0} [0] \rangle^{(1)}$$

and

$$\langle a^\dagger (\omega_1), a (\omega_2) \rangle^{(4)} = -2\alpha^2 \delta (\omega_1 - \omega_2) |G_{s_0,a_0} [\omega_1]|^2 |T_0|^2$$

(10)

(11)

The previous expressions involve Green’s functions relative to the symmetric Rydberg spinwave annihilation operator $s_0 \equiv \sum_n c_{n,0}$ as well as the term $T_0$ which accounts for the quantum interference of all possible scattering processes of two symmetric Rydberg spinwaves at all orders in $H_{dd}$. The value of $T_0$ takes into account the resummation of the series in $H_{dd}$ and it can be evaluated as the specific element $T_0 \equiv T[0,0]$ of the matrix $T[\vec{k}, \vec{k}']$ which describes the scattering of two incoming into two outgoing Rydberg spinwaves of respective wavevectors $(\vec{k}, -\vec{k})$ and $(\vec{k}', -\vec{k}')$, as considered in \[14\]. Using, e.g., diagrammatic techniques \[14\], one derives the self-consistent equation

$$T[\vec{k}, \vec{k}'] = U_{\vec{k}, -\vec{k}} + i \sum_q S_q U_{\vec{k}, -\vec{q}} T[\vec{q}, \vec{k}']$$

\hspace{1cm} (12)

where $S_q \equiv \frac{1}{2\pi} \int d\omega \ G_{s_0, -\vec{q} \cdot \vec{r}_n} [-\omega] G_{s_0, \vec{q} \cdot \vec{r}_n} [\omega]$, $s_0 \equiv \frac{1}{\sqrt{N}} \sum_n e^{-i\vec{q} \cdot \vec{r}_n} c_{n,0}$ is the Rydberg spinwave with the wavevector $\vec{q}$ and $U_{\vec{k}, -\vec{q}}$ is the Fourier transform of the interaction potential $C_{0/||\vec{p}||}$. It can be shown that for $\vec{q} \neq 0$ all Green’s functions $G_{s_0, \vec{q} \cdot \vec{r}_n} [\omega]$ coincide whence $S_q \equiv 0$ and therefore Eq. (12) becomes

$$T[\vec{k}, \vec{k}'] = U_{\vec{k}, -\vec{k}} + i S_0 U_{\vec{k}} T[0, \vec{k}'] + i S \sum_{\vec{q} \neq 0} U_{\vec{k}, -\vec{q}} T[\vec{q}, \vec{k}']$$

\hspace{1cm} (13)

$T$ can now be related to its value, denoted $\tilde{T}$ in the fictitious situation when $g = 0$, i.e. atoms are decoupled from the cavity. In that case, one indeed shows that $S_0 = S$ and therefore Eq. (13) yields

$$\tilde{T}[\vec{k}, \vec{k}'] = U_{\vec{k}, -\vec{k}} + i S \sum_q U_{\vec{k}, -\vec{q}} \tilde{T}[\vec{q}, \vec{k}']$$

\hspace{1cm} (14)

Finally, from Eqs. (13, 14), one gets

$$T[\vec{k}, \vec{k}'] = \tilde{T}[\vec{k}, \vec{k}'] + \frac{i (S_0 - S) \tilde{T}[0, 0] \tilde{T}[0, \vec{k}']} {1 - i (S_0 - S) \tilde{T}[0, 0]}$$

\hspace{1cm} (15)

and therefore $T_0 = \tilde{T}_0 / [1 - i (S_0 - S) \tilde{T}_0]$ where we defined $\tilde{T}_0 \equiv \tilde{T}[0, 0]$. When $g = 0$, Rydberg excitations cannot hop from the atom where they were created to another: the calculation of $\tilde{T}_0$ is therefore a simple twobody problem and, for a sample of volume $V$, we find

$$\tilde{T}_0 = \frac{1}{N^2} \sum_{mn} \frac{\kappa_{mn}} {1 - i \kappa_{mn} S} \approx \frac{2\pi^2} {3V} \sqrt{-1/C_0} / S$$

In principle, we can numerically compute physical quantities for arbitrary excitation numbers $(k \geq 2)$, provided that excitation exchange between atoms via the cavity mode is neglected; this approximation is valid for samples much larger than the blockade radius $(v_b \ll V)$. The calculations involve the so-called Faddeev equations and will be presented elsewhere. In the following section we present and analyze an exact analytic expression derived from the two-excitation computation above.

Transmitted light spectrum. The previous results allow us to investigate the spectrum of the light transmitted through the cavity, i.e. $S(\omega) \equiv \int d\nu \langle a^\dagger_{out} (\omega) a_{out} (\nu) \rangle = 2\gamma_c (d) \int d\nu \langle a^\dagger (\omega) a (\nu) \rangle$ \[12\]. Expanding $S(\omega)$ up to the fourth order in $\alpha$ and using Eqs. (13, 14), we find $S(\omega) \approx S^{(2)}(\omega) + S^{(4)}(\omega)$, where $S^{(2)}(\omega) = 4\gamma_c (d) \alpha^2 |G_{s_0,a_0} [0]|^2 \delta (\omega)$ while $S^{(4)}(\omega) = S^{(4)}(\omega) + S^{(4)}(\omega)$ with

$$S^{(4)}(\omega) = 4\gamma_c (d) \pi \alpha^4 |G_{s_0,a_0} [0]|^2 \delta (\omega)$$

In other words, whereas at the second order in $\alpha$ the transmitted light has the same frequency as the probe field, the fourth order spectrum comprises both an elastic contribution $S^{(4)}(\omega)$ – which partially compensates the second order contribution $S^{(2)}(\omega)$, and an inelastic contribution $S^{(4)}(\omega)$, which renders the transmitted light slightly polychromatic. Though physically expectable, this behaviour had never been reported, to our knowledge. Fig. \[6\] displays the fourth-order inelastic component $S^{(4)}(\omega)$ as a function of the frequency
Figure 2. The level structure of the Hamiltonian (4) restricted to two excitations. The structure of the doubly excited manifold is represented schematically.

\( \omega \) and the control field Rabi frequency \( \Omega_{cf} \) in: (a) resonant \( (\Delta_c = \Delta_e = \Delta_r = 0) \) and (b) detuned \( (\Delta_c = -3\gamma_e, \Delta_e = 0, \Delta_r = 0) \) configurations. We moreover assume a cloud cooperativity \( C \equiv \frac{g^2 N}{\gamma_c \gamma_e} = 5, \) and \( \gamma(c) \approx 3\gamma_e \gg \gamma(d) \). The cavity, Rydberg and intermediate state decays, respectively. In both cases, \( S^{(4)}(\omega) \) shows resonances – three for the resonant configuration (Fig. 3a), six for the generic detuned case (Fig. 3b), whose frequencies’ absolute values correspond to the three polariton eigenenergies \( \{\epsilon_k = 1, 2, 3\} \) of the Hamiltonian in the single excitation subspace, written in the frame rotating at the probe frequency

\[
\begin{pmatrix}
-\Delta_c & g\sqrt{N} & 0 \\
g\sqrt{N} & -\Delta_e & \Omega_{cf} \\
0 & \Omega_{cf} & -\Delta_r
\end{pmatrix}
\] (16)

This can be understood by noticing that \( S^{(4)}(\omega) \) corresponds to a four-photon process, in which two probe photons are absorbed by the system which subsequently reemits two photons through radiative or cavity decays via the three polariton states (see Fig. 2. 3). Three two-photon emission channels are possible, corresponding to the three polariton energies \( \epsilon_k \). For each channel \( k \), the reemitted photons’ frequencies are respectively given by \( (\omega_p \pm \epsilon_k) \), that is \( \pm \epsilon_k \) in the frame rotating at the probe frequency, which leads up to six resonances, seen in Fig. 3b). In the specific resonant case (a), \( \epsilon_1 = \epsilon_3 \) and therefore only three resonances can be identified (cf Fig. 3a). Let us note that, since \( H_{dd} \) enters Eq. (14) only via the overall factor \( T_0 \), the strength of dipole-dipole interactions does not affect the \( \omega \)-dependence of the inelastic component at fourth order but merely governs its order of magnitude.

**Conclusion.** In this Letter, we investigated the quantum optical nonlinearities induced by a cavity Rydberg EIT medium. Using the Schwinger-Keldysh contour formalism, we analytically computed field correlation functions beyond the lowest non-vanishing order in feeding, and the transmitted light spectrum. Its inelastic part appears to contain several resonances explained by a simple polaritonic picture. For simplicity, we assumed a constant feeding of the system: the formalism allows, however, for time-dependent wavepacket inputs as well. Though rarely employed in this context, the contour formalism is a powerful tool for quantum optics which could be used, e.g., to compute higher-order correlation functions of the system or thoroughly analyze subtle effects in Rydberg atomic ensembles such as thermalization [17] or phase transition [18].

This work is supported by the European Union grants SIQS (FET # 600645) and RySQ (FET # 640378), and by the « Chaire SAFRAN - IOGS Photonique Ultime ».

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Figure 3. Logarithm of the cavity transmission spectrum \( S^{(L)}(\omega) \equiv \log_{10} \left[ 2\gamma(d) \int d\nu \langle a^\dagger(\omega), a(\nu) \rangle^{(4)} \right] \) at fourth order as a function of \( \Omega_{cf} \) and the frequency (in the frame rotating at \( \omega_p \)) for: a) the resonant case \( \Delta_c = \Delta_e = \Delta_r = 0 \), b) the detuned case. The transverse curves give \( (\pm \epsilon_1, \pm \epsilon_2, \pm \epsilon_3) \) as functions of \( \Omega_{cf} \) (see main text).
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