TRK Sum Rule for Interacting Photons

Salvatore Savasta,1 Omar Di Stefano,1,2,* and Franco Nori2,3

1 Dipartimento di Scienze Matematiche e Informatiche,
Scienze Fisiche e Scienze della Terra,
Università di Messina, I-98166 Messina, Italy
2 Theoretical Quantum Physics Laboratory,
RIKEN Cluster for Pioneering Research,
Wako-shi, Saitama 351-0198, Japan
3 Physics Department, The University of Michigan,
Ann Arbor, Michigan 48109-1040, USA

Abstract

The Thomas-Reiche-Kuhn sum rule is a fundamental consequence of the position-momentum commutation relation for an atomic electron and it provides an important constraint on the transition matrix elements for an atom. Here we propose a TRK sum rule for electromagnetic fields which is valid even in the presence of very strong light-matter interactions and/or optical nonlinearities. While the standard TRK sum rule involves dipole matrix moments calculated between atomic energy levels (in the absence of interaction with the field), the sum rule here proposed involves expectation values of field operators calculated between general eigenstates of the interacting light-matter system. This sum rule provides constraints and guidance for the analysis of strongly interacting light-matter systems and can be used to test the validity of approximate effective Hamiltonians often used in quantum optics.

* corresponding author: odistefano@unime.it
Since the beginning of quantum mechanics, sum rules have proved to be very useful for understanding the general features of difficult problems. These relations, obtained by adding (sum) unknown terms, power tool for the study of physical processes [1]. Historically, the first important sum rule is found in atomic physics and concerns the interaction of electromagnetism with atoms: the Thomas-Reiche-Kuhn (TRK) sum rule [2–4]. It states that the sum of the squares of the dipole matrix moments from any energy level, weighted by the corresponding energy differences, is a constant. The TRK and analogous sum rules, like the Bethe sum rule [5], play an especially important role in the interaction between light and matter. They have widely been applied to the problems of electron excitations in atoms, molecules, and solids [6].

For an atomic electron, the TRK sum rule is a direct consequence, of the canonical commutation relation between position and momentum. It is possible to view it as a necessary condition in order not to violate this commutation relation [7]. Among the many consequences of this sum rule, it constrains the cross sections for absorption and stimulated emission [8]. It has also been shown that useful sum rules can be obtained for nonlinear optical susceptibilities [9–11]. A modified TRK sum rule for the motion of the atomic center of mass and a generalized TRK sum rule to include ions have been also obtained [12]. Extensions of the TRK sum rule to the relativistic case have been studied (see, e.g., [13, 14]). Important sum rules have also been developed in quantum chromodynamics (see, e.g., [15]).

Almost all the developed sum rules have been derived for the degrees of freedom of particles. One exception is consituted by Ref. [16], where optical sum rules have been derived for polaritons propagating through a linear medium. Here we propose a TRK sum rule for electromagnetic fields which is valid even in the presence of very strong light-matter interactions and/or optical nonlinearities [17, 18]. While the standard TRK sum rule involves dipole matrix moments calculated between atomic energy levels (in the absence of interaction with the field), the sum rule here proposed involves field operators calculated between general eigenstates of the interacting light-matter system (dressed light-matter states).

In the last years, several methods to control the strength of the light-matter interaction have been developed, and the ultrastrong coupling (USC) between light and matter has transitioned from theoretical proposals to experimental realities [17, 18]. In this new regime of quantum light–matter interaction, beyond weak and strong coupling, the coupling strength becomes comparable to the transition frequencies in the system, or even higher [deep strong...
coupling (DSC)] [19–22]. In the USC and DSC regimes, approximations widely employed in quantum optics break down [23], allowing processes that do not conserve the number of excitations in the system (see, e.g., [24–28]). The non-conservation of the excitation number gives rise to a wide variety of novel and unexpected physical phenomena in different hybrid quantum systems [25, 29–49]. As a consequence, all the system eigenstates, dressed by the interaction, contain different numbers of excitations. Much research on these systems has dealt with understanding whether these excitations are real or virtual, how they can be probed or extracted, how they make possible higher-order processes even at very low excitation densities, and how they affect the description of input and output for the system [17].

The eigenstates of these systems, including the ground state, can display a complex structure involving superposition of several eigenstates of the non-interacting subsystems [17, 18, 50], and can be difficult to calculate. As a consequence, a number of approximation methods have been developed [51, 52]. Moreover, the output field correlation functions, connected to measurements, depend on these eigenstates (see, e.g., Ref. [39, 53]). Hence sum rules providing general guidance and constraints can be very useful to test the validity of the approximations. This general sum rule can also be used to test the validity of effective Hamiltonians often used in quantum optics and cavity optomechanics [49, 54, 55]. In addition, this generalized TRK sum-rule applies to the broad emerging field of nonperturbative light-matter interactions, including several settings and subfields, as cavity and circuit-QED [17], collective excitations in solids [56], optomechanics [54], photochemistry and QED chemistry [50, 57].

**Sum rule for interacting photons. —** A key property used for the derivation of the TRK sum rule is that the commutator between the electron coordinate and the electronic Hamiltonian does not depend on the electronic potential, which is a function of the coordinate only, and hence it is universal. Considering for simplicity a single electron 1D system, if \( \hat{q} \) is the electron coordinate and \( \hat{H}_{at} = \hat{p}^2/2m + V(\hat{q}) \) is the electronic Hamiltonian: 

\[
[\hat{q}, \hat{H}_{at}] = [\hat{q}, \hat{p}^2/2m] = i(h/m)\hat{p}.
\]

In the Coulomb gauge, the (transverse) vector potential \( \mathbf{A} \) represents the field coordinate, while its conjugate momentum \( \Pi \) is proportional to the transverse electric field:

\[
\Pi(x, t) = -\varepsilon_0 \mathbf{E}(x, t) = \varepsilon_0 \mathbf{A}(x, t).
\]
A general feature of the light-matter interaction Hamiltonians derived from the *minimal coupling replacement* (as for the Coulomb gauge) is that the momenta of the matter system are coupled *only* to the field coordinate. We can express the total light-matter quantum Hamiltonian as

\[ \hat{H} = \hat{H}_F + \hat{H}_M + \hat{H}_I, \]

where the first two terms on the r.h.s. are the field and matter system free Hamiltonians, and the third describes the light-matter interaction. Using Eq. (1) and the Heisenberg equation \( i\hbar \dot{\hat{A}} = [\hat{A}, \hat{H}] \), we obtain the relation

\[ i\hbar \Pi = \varepsilon_0[\hat{A}, \hat{H}] = \varepsilon_0[\hat{A}, \hat{H}_F], \]  

where the second equality follows from \([\hat{A}, \hat{H}_I] = 0\), which holds, e.g., in the Coulomb gauge. For simplicity, we consider the case of a quasi 1D electromagnetic resonator of length \( L \), so that the expression for the electric-field operator can be simplified to \( \hat{E}(r, t) \rightarrow \tilde{s} \hat{E}(x, t) \), where \( \tilde{s} = y/|y| \). The vector potential (as well as the electric field operator) can be expanded in terms of photon creation and destruction operators as

\[ \hat{A}(x, t) = \sum_m A_m(x) \hat{a}_m e^{-i\omega_m t} + \text{h.c.} \]  

and

\[ \hat{E}(x, t) = \sum_m E_m(x) \hat{a}_m e^{-i\omega_m t} + \text{h.c.}, \]

where \( A_m(x) = [\hbar/(2\omega_m \varepsilon_0 A)]^{1/2} u_m(x) \), and \( E_m(x) = i\omega_m A_m(x) \). Here, \( AL \) is the resonator volume, the subscript \( m \) labels a generic mode index with frequency \( \omega_m \), and \( u_m(x) \) are the normal modes of the field chosen as real functions. For example, imposing the vanishing of the electric field at the two end walls at \( x = \pm L/2 \) of the cavity, \( u_m(x) = (1/\sqrt{L}) \sin k_m(x + L/2) \), where \( k_m = \pi m/L \).

Let us now consider the matrix elements of the operators in Eq. (2) between two generic eigenstates \( |\psi_i\rangle \) of the total Hamiltonian \( \hat{H} \). We obtain

\[ \Pi_{ij} = i\varepsilon_0 \omega_{ij} A_{ij}, \]  

where \( \omega_{ji} = \omega_j - \omega_i \) and we used the notation \( O_{ij} = \langle \psi_i | \hat{O} | \psi_j \rangle \). Here and in the following, \( j = 0 \) indicates the system ground state, and the energy levels are ordered according to their energy: \( j > i \) if \( \omega_j > \omega_i \). We now multiply both sides of Eq. (3) by \( u_m(x) \) and integrate over \( x \). By defining \( \hat{Q}^{(m)} = (\hat{a}_m + \hat{a}_m^\dagger)/\sqrt{2} \), and \( \hat{P}^{(m)} = i(\hat{a}_m^\dagger - \hat{a}_m)/\sqrt{2} \), we obtain the corresponding relation for the individual modes:

\[ \omega_m P^{(m)}_{ij} = i\omega_{ij} Q^{(m)}_{ij}. \]  

It is worth noticing that, in the limit when the interaction vanishes, \( |P^{(m)}_{ij}| = |Q^{(m)}_{ij}| \), and Eq. (4) can easily be verified analytically. When the interaction becomes relevant, so that the system eigenstates differ from the harmonic spectrum for free fields, the ratio between the
two quadratures can be very different from 1 and can be determined by the only knowledge of the energy spectrum, independently on the specific interacting system. Equation (4) is the first result of this work. It shows that the ratio between the two field quadratures is uniquely determined by the energy spectrum. The two quadratures can display very different matrix elements when the interaction with the matter field changes significantly the energy levels of the interacting systems, as it occurs in the USC and DSC regimes.

Let us now consider the commutator between the mode coordinate and its conjugate momentum:

\[ i = \left[ \hat{Q}^{(m)}, \hat{P}^{(m)} \right] = \frac{1}{i\hbar\omega_m} \left[ \hat{Q}^{(m)}, \left[ \hat{Q}^{(m)}, \hat{H}_F \right] \right], \tag{5} \]

where we used \( \omega_m \hat{P}^{(m)} = \dot{\hat{Q}}^{(m)} \), and \( \left[ \hat{Q}^{(m)}, \hat{H}_F \right] = \left[ \hat{Q}^{(m)}, \hat{H}_F \right] \). Developing the double commutator, considering its matrix elements between two generic eigenstates of the total Hamiltonian \( \hat{H}_F \), and inserting the identity operators (\( \hat{I} = \sum_k |\psi_k\rangle\langle\psi_k| \)), we obtain the following relation

\[ \sum_k \omega_{k,i} + \omega_{k,j} \omega_m \hat{Q}_{i,k}^{(m)} \hat{Q}_{k,j}^{(m)} = \delta_{i,j}, \tag{6} \]

which reduces (choosing \( j = i \)) to the TRK sum rule for interacting fields:

\[ 2 \sum_k \frac{\omega_{k,i}}{\omega_m} |\hat{Q}_{i,k}^{(m)}|^2 = 1. \tag{7} \]

By using Eq. (4), Eq. (7) can be also expressed in terms of the momenta matrix elements:

\[ 2\omega_m \sum_k |\hat{P}_{i,k}^{(m)}|^2 / \omega_{k,i} = 1. \]

Formally, it coincides with the TRK sum rule for atoms; however, in Eq. (7) the matrix elements of the field-mode coordinate replace the atomic electric-dipole matrix elements. An important difference is that the atomic TRK sum rule [58] considers atomic energy eigenstates, calculated in the absence of interaction with the field. On the contrary, this sum rule is very general, since it holds in the presence of interactions with arbitrary matter systems, every time the interaction occurs via the field coordinate (e.g., Coulomb gauge). We also observe that Eq. (7) describes a collection of sum rules, one for each field mode \( m \). Actually, following the same reasoning which led us to Eq. (7), a generalized atomic TRK sum rule for atoms strongly interacting with the electromagnetic field [analogous to Eq. (7)] can be easily obtained. The resulting atomic generalized TRK sum rule, formally coincides with the standard one, with the only difference that all the expectation values are calculated using the eigenstates of the total light-matter system:

\[ 2m \sum_k \omega_{k,j} |\hat{x}_{k,j}|^2 = 1, \tag{8} \]
where we considered a single electron system, and $x_{k,j}$ is the expectation value of the position operator between two dressed states.

**Quantum Rabi model.** — The quantum Rabi Hamiltonian, describes the dipolar coupling between a two-level atom and a single mode of the quantized electromagnetic field. Recently, it has been shown [59] that the correct (satisfying the gauge principle) quantum Rabi Hamiltonian in the Coulomb gauge

$$\hat{H}_C = \hbar \omega_c \hat{a}^\dagger \hat{a} + \frac{\hbar \omega_0}{2} \left\{ \hat{\sigma}_z \cos \left[ 2\eta (\hat{a} + \hat{a}^\dagger) \right] \right\} + \hat{\sigma}_y \sin \left[ 2\eta (\hat{a} + \hat{a}^\dagger) \right], \tag{9}$$

strongly differs from the standard model (see also Refs [60–62] for gauge issues in the USC regime). Here, $\omega_c$ is the resonance frequency of the cavity mode, $\omega_0$ is the transition frequency of a two-level atom, $\hat{a}$ and $\hat{a}^\dagger$ are the destruction and creation operators for the cavity field, while the qubit degrees of freedom are described by the Pauli operators $\hat{\sigma}_i$. When the normalized coupling strength is small ($\eta \ll 1$), considering only first order contributions in $\eta$, the standard interaction term $\hbar \omega_0 \eta (\hat{a} + \hat{a}^\dagger) \hat{\sigma}_y$ is recovered. If the system is prepared in its first excited state, the photodetection rate for cavity photons is proportional to $|P_{1,0}|^2$ (see Ref. [53, 62]). Figure 1(a) displays this quantity (red continuous curve) as well as $|Q_{1,0}|^2$ (dashed blue) versus the normalized coupling $\eta$, calculated after the numerical diagonalization of Eq. (9). The two quantities are equal only at negligible coupling. However, in agreement with Eq. (4), the numerically calculated $(\omega_{1,0}^2/\omega_c^2)|Q_{1,0}|^2$ (black dashed curve) coincides with $|P_{1,0}|^2$. In contrast, the JC model $[\hat{H}_{JC} = \hbar \omega_c \hat{a}^\dagger \hat{a} + \hbar \omega_0/2 \hat{\sigma}_z + \hbar \eta \omega_c (\hat{a} \hat{\sigma}_+ + h.c.)]$, violates Eq. (4) providing coupling-independent values $|Q_{1,0}|^2 = |P_{1,0}|^2$ [the horizontal line in Fig. 1(a)].

In order to understand how the sum rule in Eq. (7) applies to the quantum Rabi model, we calculate partial sums with an increasing number of states. Specifically, we calculate $\sum_{j=1}^{N} \mathcal{F}_{0j}$, where $\mathcal{F}_{0j} = 2(\omega_{j,0}/\omega_c)|Q_{0,j}|^2$. Differently from the JC, the quantum Rabi model does not conserve the excitation number. Therefore, expectation values like $Q_{0,j}$ (and hence $\mathcal{F}_{0,j}$) can be different from zero also for $j > 2$. Figure 1(b) displays such partial sums as a function of the number of levels included, obtained for different values of $\eta$. For small values ($\eta = 0.01$) only the two lowest excited levels contribute to the sum with approximately equal weights, in good agreement with the JC model. For $\eta = 0.2$ only two transitions contribute to the sum rule; however the second transition provides a larger contribution to the sum. For $\eta = 0.5$, the contribution of the lowest energy transition become smaller, while
Figure 1. (a) $\mathcal{P}$–$\mathcal{Q}$ relation: calculation of $|\mathcal{P}_{1,0}|^2$ (proportional to the photodetection rate for cavity photons) (red continuous curve) and of $|\mathcal{Q}_{1,0}|^2$ (dashed blue) versus the normalized coupling $\eta$. (b) TRK sum rule for interacting fields: partial sums $\sum_{j=0}^{N} F_{0j}$ as function of the number $N$ of levels included for different normalized coupling rates $\eta$. Inset: energy spectrum for the first energy levels $\omega_{k,0}$ versus the normalized coupling strength.
\( \mathcal{F}_{02} = 0 \), owing to the parity selection rule. Notice that, at \( \eta = \eta_{cr} \simeq 0.44 \) there is a crossing between the levels 2 and 3 [see inset in Fig. 1(b)], so that, for \( \eta > \eta_{cr} \), state \( |2 \rangle \) has the same parity of state \( |0 \rangle \). It is sufficient to include \( \mathcal{F}_{03} \) to approximately satisfy the sum rule. For \( \eta = 1 \), \( \mathcal{F}_{0,1} \) is very small and \( \mathcal{F}_{1,2} = 0 \). In this case the sum rule is satisfied mainly with the contributions \( \mathcal{F}_{0,j} \) with \( 3 \leq j \leq 6 \). Finally, for very high values of the normalized coupling strength (\( \eta = 1.8 \)) only one contribution (\( \mathcal{F}_{0,3} \)) becomes relevant. This effect is due to the light-matter decoupling [62] which occurs at very high values of \( \eta \), where the system ground state \( |0 \rangle \) is well approximated by \( |g,0 \rangle \), then \( |1 \rangle \simeq |e,0 \rangle \), \( |2 \rangle \simeq |e,1 \rangle \), \( |3 \rangle \simeq |g,1 \rangle \), and so on: the higher energy levels are of the kind \( \simeq |g(e),n > 1 \rangle \). This explains why for \( \eta = 1.8 \) the only significant contribution to the sum is \( \mathcal{F}_{0,3} \). These behaviours of the partial sums and of the terms \( \mathcal{F}_{i,j} \) are closely connected to accessible experimental features, as explicitly shown in the example below.

**Nonlinear electromagnetic resonator.** — As a further test, we analyze a single-mode nonlinear optical system described by the following effective Hamiltonian

\[
\hat{H} = \hbar \omega_c \hat{a}^\dagger \hat{a} + \eta \hbar \omega_c \left( \hat{a} + \hat{a}^\dagger \right)^3 + \frac{\eta}{10} \hbar \omega_c \left( \hat{a} + \hat{a}^\dagger \right)^4.
\]

(10)

Here \( \hat{H}_F = \hbar \omega_c \hat{a}^\dagger \hat{a} \), while the nonlinear terms are assumed to arise from the dispersive interaction with some material system [63]. Note that the nonlinear terms in Eq. (10) commutates with the field coordinate \( \hat{Q} = (\hat{a} + \hat{a}^\dagger) / \sqrt{2} \), hence Eqs. (4) and (7) holds. In contrast, the presence of a standard self-Kerr term \( \propto \hat{a}^\dagger 2 \hat{a}^2 \) (see, e.g., [64]) would violate them. The inset in Fig. 2 shows the anharmonic energy spectrum \( \omega_{k,0} \) as a function of \( \eta \). Figure (2) displays the partial sums \( \sum_{j=1}^{N-1} \mathcal{F}_{0j} \) as versus the number of included levels, calculated for different values of \( \eta \). Increasing the anharmonicity coefficient \( \eta \), the number of contributions in the sum increases at the expense of the contribution \( \mathcal{F}_{01} \) of the lowest energy transition. This behaviour is closely connected with accessible experimental features which can be observed, e.g., in linear transmission spectra. For a two-port (equally coupled to the external modes) nonlinear resonator, the transmission spectrum (see Appendix A) can be written as

\[
T(\omega) = \omega^2 \left| \sum_k \frac{\Gamma_{k,0}/\omega_{k,0}}{\omega_{k,0} - \omega - i\Gamma_k} \right|^2,
\]

(11)

where the radiative decay rates are \( \Gamma_{k,j} = 2\pi g^2(\omega_{k,j})|\mathcal{Q}_{k,j}|^2 \), \( \Gamma_k = \sum_{j<k} \Gamma_{k,j} \), and we assumed an ohmic coupling with the external modes (\( g^2(\omega) \propto \omega \)). When the anharmonicity is switched off (\( \eta = 0 \)), \( \Gamma_{k,0} \propto \mathcal{F}_{0k} = 0 \) for \( k \neq 1 \), and the transmission spectrum presents
Figure 2. (a) TRK sum rule for a single-mode nonlinear system: partial sums $\sum_{j=0}^{N} F_{0j}$ versus the number ($N$) of levels included for different normalized coupling strengths $\eta$. Inset: anharmonic energy spectrum $\omega_{k,0}$ versus $\eta$. (b) Transmission spectrum $T(\omega)$ for a two-port nonlinear resonator for $\eta = 0.12$. The inset shows the integrated lines for two values of $\eta$. 
a single peak at $\omega = \omega_c$ [see Fig. 2(b)]. When $\eta \neq 0$, $\Gamma_{k,0} \propto F_{ok} \neq 0$, and the transmission spectrum in Fig. 2 evolves accordingly (the blue-continuous curve show the spectrum calculated for $\eta = 0.12$). By integrating the individual spectral lines in Eq. (11), we obtain for each line a contribution $\simeq \pi \Gamma_{k,0}^2 / \Gamma_k$, which is approximately proportional to $F_{ok}$ in the sum (notice that $\Gamma_k \sim k \Gamma_1$). The inset in Fig. 2 shows the integrated lines for two values of $\eta$.

**Discussion.**— The relations in Eq. (4) and Eq. (7) are very general. So far we applied them to single-mode fields, however they are also valid in the presence of (even interacting) multi-mode fields (see, e.g., [65, 66]) and systems including several dipoles (see, e.g., [67, 68]). In Appendix B we discuss an example where two modes of different frequency interact via a qubit [43]. The TRK sum rule for interacting photons here proposed can also be useful to analyze general quantum nonlinear optical effects (see, e.g., [27, 69–71]).

Approximate Hamiltonians and effective models can violate one of the two relations here proposed, which are intimately tied to the indeterminacy principle. Such a violation indicates that the model may miss some relevant physics. For example, we have shown that the JC model violates the relation in Eq. (4). An additional example of a model violating this relation is provided by the well-known and widely employed cavity optomechanical interaction Hamiltonian $\hbar g \hat{a}^{\dagger} \hat{a} \hat{b}^{\dagger} (\hat{b} + \hat{b}^{\dagger})$ (here $\hat{b}$ is the destruction operator for the mechanical oscillator) [72]. On the contrary, the interaction Hamiltonian obtained by a microscopic model [54] $\hbar g (\hat{a}^{\dagger} + \hat{a})^{2} (\hat{b} + \hat{b}^{\dagger})$, satisfies both of these relations [Eqs (7), (8)]. It turns out that such interaction Hamiltonian, in addition to the standard optomechanical effects, also describes the dynamical Casimir effect [49, 55].

An interesting feature of the relations proposed here is that they hold in the presence of light-matter interactions of arbitrary strength. Moreover, the obtained sum rule is useful for the analysis of strongly interacting light-matter systems, especially when exact eigenstates are not available. These relations in Eq. (4) and Eq. (7) can provide constraints and a guidance in the development of effective Hamiltonians in quantum optics and cavity optomechanics. Finally, following the same reasoning leading to Eq. (7), we also obtained a generalized TRK sum rule involving transitions between the *total* light-matter energy eigenstates [Eq. (8)].
ACKNOWLEDGMENTS

F.N. is supported in part by the: MURI Center for Dynamic Magneto-Optics via the Air Force Office of Scientific Research (AFOSR) (FA9550-14-1-0040), Army Research Office (ARO) (Grant No. Grant No. W911NF-18-1-0358), Japan Science and Technology Agency (JST) (via the Q-LEAP program, and the CREST Grant No. JPMJCR1676), Japan Society for the Promotion of Science (JSPS) (JSPS-RFBR Grant No. 17-52-50023, and JSPS-FWO Grant No. VS.059.18N), the RIKEN-AIST Challenge Research Fund, the Foundational Questions Institute (FQXi), and the NTT PHI Laboratory. S.S. acknowledges the Army Research Office (ARO) (Grant No. W911NF1910065).

Appendix A: Linear response theory and transmission of a nonlinear optical system

This section provides a pedagogical derivation of the dressed master equation in the Schrödinger picture can be written as [73],

\[ \dot{\hat{\rho}}(t) = -i \left[ \hat{H}_S, \hat{\rho}(t) \right] + \mathcal{L}\hat{\rho}(t), \tag{A1} \]

where

\[ \hat{H}_S = \sum_k \omega_k |k\rangle \langle k|, \tag{A2} \]

with the Lindbladian superoperator defined by

\[ \mathcal{L}\hat{\rho}(t) = \sum_i \sum_{j,k<j} \left\{ \Gamma_i^{jk} n(\omega_{jk}, T_i) \mathcal{D}[[j]\langle k|] \hat{\rho}(t) + \Gamma_i^{jk} [1 + n(\omega_{jk}, T_i)] \mathcal{D}[[k]\langle j|] \hat{\rho}(t) \right\}. \tag{A3} \]

The equation above includes the thermal populations

\[ n(\Delta_{jk}, T_i) = \left[ \exp \{ \omega_{jk}/k_BT_i \} - 1 \right]^{-1}, \tag{A4} \]

and the damping rates

\[ \Gamma_i^{jk} = 2\pi g_i(\omega_{jk}) |\alpha_i(\Delta_{jk})|^2 |C_{jk}|^2. \tag{A5} \]

with \( i = \{L, R\} \) indicating the input-output ports, \( g(\Delta_{jk}) \) being the reservoir density of states, \( \alpha(\Delta_{jk}) \) the system-reservoir coupling strength, and the generic dissipator operator

\[ \mathcal{D}[\hat{O}] \hat{\rho} = \frac{1}{2} \left( 2\hat{O}\hat{\rho}\hat{O}^\dagger - \hat{\rho}\hat{O}^\dagger\hat{O} - \hat{O}^\dagger\hat{O}\hat{\rho} \right). \tag{A6} \]
At $T = 0$, being $n(\Delta_{jk}, T_i) = 0$, we obtain
\begin{equation}
\mathcal{L}\hat{\rho} \bigg|_{T=0} = \mathcal{L}_0\hat{\rho} = \sum_i \sum_{j<k} \left\{ \Gamma^{jk}_i D \right\} \hat{\rho} \right).
\end{equation}

Considering also a driving field entering from the left port whose Hamiltonian is given by:
\begin{equation}
\hat{H}_d = i\hat{X} \int d\omega g_L(\omega)(\hat{b}_\omega - \hat{b}_\omega^\dagger),
\end{equation}
where $\hat{X} = \hat{a} + \hat{a}^\dagger$, Eq. (A1) becomes
\begin{equation}
\dot{\hat{\rho}}(t) = -i \left[ \hat{H}_S + \hat{H}_d, \hat{\rho}(t) \right] + \mathcal{L}_0\hat{\rho}(t).
\end{equation}

We assume that the light field from the left port is coherent with driving frequency $\omega$: $\langle \hat{b}_\omega \rangle = \beta_L(\omega) \exp[-i\omega t]$. Retaining only the terms depending linearly from the input field and using Eqs. [(A1), (A7), (A8)], assuming $\rho_{n0}(t) = \rho_{n0} \exp[-i\omega t]$ (i.e., oscillating resonantly with the driving field), and using the rotating wave approximation, we obtain (to first order in the field)
\begin{equation}
\rho^{(1)}_{n0} = \frac{i g_L(\omega)\beta_L(\omega)X_{n0}}{(\omega - \omega_{n0}) + i \sum_i \sum_{k<n} \Gamma^{n,k}_i},
\end{equation}
where, being $T = 0$, only the ground state is populated in the absence of interaction ($\rho^{(0)}_{00} = 1$). In order to calculate the transmitted signal that can be experimentally detected, we consider a system constituted by an $LC$-oscillator coupled to a transmission line and use the input-output relations [62] for the positive frequency component of the output (input) vector potential operator defined as
\begin{equation}
\hat{\phi}^{\pm}_{\text{out(in)}}(t) = \Lambda \int_0^\infty \frac{d\omega}{\sqrt{\omega}} \hat{\phi}^{\text{out(in)}}_{\omega}(t),
\end{equation}
where, for the sake of simplicity, we disregarded the spatial dependence, and $\Lambda = \sqrt{\hbar Z_0/4\pi}$, with $Z_0$ the impedance of the in-out transmission line(s). In addition, we consider two distinct ports for the input ($L$) and the output ($R$) [for simplicity we assume $g_L(\omega) = g_R(\omega) = g(\omega)$] and we have for the output voltage operator [62] $\hat{V}^{(R)+}_{\text{out}}(t) = \hat{\phi}^{(R)+}_{\text{out}}(t)$:
\begin{equation}
\hat{V}^{(R)+}_{\text{out}}(t) = -2\pi \Lambda \sum_j \frac{g(\omega_{j0})}{\sqrt{\omega_{j0}}} X_{0j} \hat{P}_{0j}(t),
\end{equation}
which can be expressed as
\begin{equation}
\hat{V}^{(R)+}_{\text{out}}(t) = -K \hat{V}^+(t),
\end{equation}
\end{equation}
where
\[ \hat{V}^+ = \Phi_{zpf} \sum_j X_{0j} \hat{P}_{0j}(t). \] (A14)

Assuming \( g(\omega) = G \sqrt{\omega} \), the constants \( K \) and \( \Phi_{zpf} \) satisfy the relation
\[ \frac{K \Phi_{zpf}}{\Lambda} = 2\pi G. \] (A15)

Using Eq. (A11), we have for the mean value of the input sent through the port (L)
\[ \langle \hat{V}_{in}^{(L)+}(t) \rangle = \langle \phi_{in}^{(L)+}(t) \rangle = -i \Lambda \sqrt{\omega} \beta_{L}(\omega), \] (A16)
where we assumed a coherent drive input at frequency \( \omega \):
\[ \langle \hat{b}_{L'}(t) \rangle = \beta_{L}(\omega) \delta(\omega' - \omega). \]
Considering the linear response only, the projection operator oscillates at the frequency \( \omega \) of the drive, \( \hat{P}_{0j}(t) = -i \omega \hat{P}_{0j}(t) \), using Eqs. \[(A13), (A14)]\, the mean value for the output is
\[ \langle \hat{V}_{out}^{(R)+}(t) \rangle = iK\Phi_{zpf} \omega \sum_j X_{0j} \rho_{j0}(t), \] (A17)
where \( \hat{\rho} \) is the density matrix and we used the relation \( \langle \hat{P}_{0j}(t) \rangle = \rho_{j0}(t) \). Using Eqs. \[(A15), (A17), (A16)]\, we can calculate the transmission coefficient \( T(\omega) \) due to the signal detected from the port (R) when a driving field is sent through the port (L) as
\[ T(\omega) = \left| \frac{\langle \hat{V}_{out}^{(R)+}(t) \rangle}{\langle \hat{V}_{in}^{(L)+}(t) \rangle} \right|^2 = \omega^2 \left| \sum_j \frac{\Gamma_{j0} \omega_{j0}}{(\omega - \omega_{j0}) + i \sum_i \sum_{k<n} \Gamma_{nk}} \right|^2, \] (A18)
where \( \Gamma_{j0} = 2\pi |g(\omega_{j0})|^2 |X_{j0}|^2 \).

Appendix B: Frequency conversion in ultrastrong cavity QED

In this section we analyze the TRK sum rule for interacting photons in a three-component system constituted by two single-mode resonators ultrastrongly coupled to a single superconducting flux qubit. This coupling can induce an effective interaction between the fields of the two resonators. Using suitable parameters for the three components, the system provides a method for frequency conversion of photons which is both versatile and deterministic. It has been shown that it can be used to realize both single and multiphoton frequency conversion processes [43]. The system Hamiltonian is
\[ \hat{H} = \hbar \omega_a \hat{a}^\dagger \hat{a} + \hbar \omega_b \hat{b}^\dagger \hat{b} + \frac{\hbar \omega_0}{2} \hat{\sigma}_z + \hbar \left[ g_a \left( \hat{a} + \hat{a}^\dagger \right) + g_b \left( \hat{b} + \hat{b}^\dagger \right) \right] \left[ \cos(\theta) \hat{\sigma}_x + \sin(\theta) \hat{\sigma}_z \right], \] (B1)
Figure 3. Energy spectrum obtained from the numerical diagonalization of Eq. (B1). (a) Lowest normalized energy levels versus the qubit frequency. (b) Enlarged view of the spectrum inside the rectangle in (a) showing the presence of an avoided level crossing. Parameters are given in the text.

where \((\hat{a}, \omega_a, g_a)\) and \((\hat{b}, \omega_b, g_b)\) describe the photon operator, the frequency mode, and the coupling with the qubit for the two resonators. The angle \(\theta\) encodes the qubit flux offset which determines parity symmetry breaking. A zero flux offset implies \(\theta = 0\). Figure 3(a) displays the lowest normalized energy levels \((\omega - \omega_g)/\bar{\omega}_0\) (we indicated with \(\hbar \omega_g\) the ground state energy) versus the qubit frequency \(\omega_0/\bar{\omega}_0\) obtained diagonalizing numerically the Hamiltonian in Eq. (B1). We used the parameters \(\omega_a = 3\bar{\omega}_0, \omega_b = 2\bar{\omega}_0, \theta = \pi/6, g_a = g_b = 0.2 \bar{\omega}_0\), where \(\bar{\omega}_0\) is a reference point for the qubit frequency. Notice that the two resonators are
Figure 4. TRK sum rule for interacting photons in the three-component system described by the Hamiltonian in Eq. (B1). (a) Partial sum rules $\sum_{j=1}^{N} F_{0j}^a$ relative to the first resonator and (b) $\sum_{j=1}^{N} F_{1j}^b$ relative to second resonator for different values of levels $N$. The black segmented line describes the zero detuning case $\delta = 0$, while the dashed blue segmented lines the case $\delta = (\omega_0 - \bar{\omega}_0) / \bar{\omega}_0 = -6 \times 10^{-3}$. Parameters are given in the text.

set in order that their resonance frequencies satisfy the relationship $\omega_a = \omega_b + \bar{\omega}_0$. The first excited level is a line with slope $\simeq 1$, corresponding to the approximate eigenstate $|\psi_1\rangle \simeq |0,0,e\rangle$, where the first two entries in the ket indicate the number of photons in resonator $a$ and $b$ respectively, while the third entry indicates the qubit state. The second
excited level is a horizontal line corresponding to the eigenstate $|\psi_2\rangle \simeq |0,1,g\rangle$, the next two lines on the left of the small rectangle in Fig. 3(a) (for values of $\omega_0/\bar{\omega}_0$ before the apparent crossing), correspond to the states $|\psi_3\rangle \simeq |0,1,e\rangle$ and $|\psi_4\rangle \simeq |1,0,g\rangle$. The apparent crossing in the rectangle is actually an avoided level crossing, as can be inferred from the enlarged view in Fig. 3(b). It arises from the hybridization of the states $|0,1,e\rangle$ and $|1,0,g\rangle$ induced by the counter-rotating terms in the system Hamiltonian. The resulting eigenstates can be approximately written as

\[
|\psi_3\rangle \simeq \cos \theta |0,1,e\rangle - \sin \theta |1,0,g\rangle \\
|\psi_4\rangle \simeq \sin \theta |0,1,e\rangle + \cos \theta |1,0,g\rangle.
\] (B2)

The mixing is maximum when the level splitting is minimum (at $\omega_0/\bar{\omega}_0 \simeq 1.056$). In this case $\theta = \pi/4$.

It has been shown [43] that this effective coupling can be used to transfer a quantum state constituted by an arbitrary superposition of zero and one photon in one resonator (e.g., $a$), to a quantum state corresponding to the same superposition in the resonator at frequency $\omega_b$.

This system represents an interesting example of two interacting optical modes (with the interaction mediated by a qubit). In order to understand how the sum rule in Eq. (7) applies to such a system, we investigate its convergence, calculating partial sum rules for the two modes. Figure 4 shows $\sum_{j=0}^{N} F^a_{0j}$ (a) and $\sum_{j=1}^{N} F^a_{1j}$ (b) for different values of $N$. The black line describes the zero detuning case, while the dashed blue line the case $\delta = (\omega_0 - \bar{\omega}_0)/\bar{\omega}_0 = -6 \times 10^{-3}$. The results in Fig. 4(a) can be understood observing that $F^a_{0j} \propto |\langle 0|\hat{a} + \hat{a}^\dagger |j\rangle|^2$. Since $|0\rangle \simeq |0,0,g\rangle$, $|1\rangle \simeq |0,0,e\rangle$, $|2\rangle \simeq |0,1,g\rangle$, $|3\rangle$ and $|4\rangle$ are provided in Eq. (B2), it is easy to obtain $F^a_{01} \simeq F^a_{02} \simeq 0$, $F^a_{03} \propto \sin^2 \theta$, and $F^a_{04} \propto \cos^2 \theta$, in agreement with the results in Fig. 4(a). Notice that for $\delta = 0$, it results in $\theta = \pi/4$, and hence $F^a_{03} \simeq F^a_{04}$. A similar analysis can be carried out for the results in Fig. 4(b).

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