A Formalism Useful to Study Beyond Squeezing in Non-linear Quantum Optics

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Abstract

A general formalism is given in quantum optics within a ring cavity, in which a non-linear material is stored. The method is Feynman graphical one, expressing the transition amplitude or S-matrix in terms of propagators and vertices. The propagator includes the additional damping effect via the non-linear material as well as the reflection and penetration effects by mirrors. Possible application of this formalism is discussed, in estimating the averaged number of produced photons, Husimi function, and the observables to examine beyond the squeezing mechanism of photons.
1 Introduction

In optics, laser technology has developed extensively and is now able to elucidate the quantum behavior of photons. That is, we can investigate quantum optics (QO) or quantum electrodynamics (QED) by using laser technologies. See for example \[1\]. It is amazing that there exist transparent materials, having non-linear dielectric constants, such as Silica, BBO (β-Barium Borate), KTP (Potassium Titanyl Phosphate), LN (Lithium Niobate), KDP (Potassium Dideuterium Phosphate) \(e.t.c\). For such non-linear materials the displacement vector (the electric flux density) \(D\) is not linearly proportional to the electric field \(E\), but we have

\[
D = \varepsilon_0(1 + \chi^{(1)})E + P_{NL}, \quad H = B/\mu_0, \text{ and} \\
(P_{NL})_i = \varepsilon_0 \sum_{i,j,k,\cdots} (\chi^{(2)}_{ijk}E_jE_k + \chi^{(3)}_{ijkl}E_jE_kE_l + \chi^{(4)}_{ijklm}E_jE_kE_lE_m + \cdots),
\]

where \(P_{NL}\) is the non-linear polarization induced by the electric field, \(\varepsilon_0\) and \(\mu_0\) are the dielectric constant and the magnetic permeability of the vacuum, respectively, and \(\chi^{(n)}\) \((n = 1, 2, \cdots)\) are dielectric constants.

The tensor structure with indices \((i,j,\cdots)\) is important for an individual material, but here we ignore it for simplicity.

Then, the action of QO or QED in the material reads

\[
S_{QO} = \frac{1}{2} \int dt \, d^3x \, (E \cdot D - H \cdot B) \\
= \frac{1}{2} \int dt \, d^3x \, \left\{ \varepsilon_0 (E^2 + \chi^{(1)}E^2 + \chi^{(2)}E^3 + \chi^{(3)}E^4 + \chi^{(4)}E^5 + \cdots) - H \cdot B \right\}
\]

which is non-linear in \(E\). In this way the non-linear dielectric constant \(\chi^{(n)}\) introduces additional interactions to the QO (or QED) Lagrangian density,

\[
\mathcal{L}_{QO}^{(n)} = \frac{1}{2} \varepsilon_0 \chi^{(n)}E(t,x)^{n+1}.
\]

The electric field is composed of the dominant classical part \(E_L\), coming from the intense laser beam, and the quantum part \(\hat{E}_q\) describing the creation and annihilation of photons:

\[
E(t,x) = E_L(t,x) + \hat{E}_q(t,x), \quad \text{where} \quad E_L(t,x) = E_L e^{-i(\omega_L t - k_L \cdot x)} + E_L^\dagger e^{i(\omega_L t - k_L \cdot x)},
\]

\[
\hat{E}_q(t,x) = -\partial_t \hat{A}_q(t,x) = \int d^3k \frac{\hbar}{(2\pi)^3} \varepsilon(i\omega) \left( \hat{a}(k)e^{-i(\omega t - k \cdot x)} - \hat{a}(k)^\dagger e^{i(\omega t - k \cdot x)} \right),
\]

where \(\hat{A}_q\) is the vector potential, \(\hat{a}(k)\) and \(\hat{a}(k)^\dagger\) are, respectively, annihilation and creation operators of photon with a wave vector \(k\), and \(\varepsilon = \varepsilon_0(1 + \chi^{(1)})\). Here we assume a simple dispersion relation, \(\omega = \omega(k) = c'|k|\), with the light velocity \(c' = 1/\sqrt{\varepsilon\mu_0}\) in the non-linear material.

\(^1\)In reality, the vector potential, creation and annihilation operators have dependence on the direction of polarization vectors, but these tensor structure is also ignored here.
Now, we understand that the intense laser beam can create or annihilate photons by the interactions, \( L_{QO}^{(n)} \) \((n = 2, 3, 4, \cdots)\), proportional to \( \chi^{(n)} \),
\[
L_{QO}^{(n)} = \frac{1}{2} (n + 1) \epsilon_0 \chi^{(n)} E_L(t, x) \hat{E}_q(t, x)^n.
\]
At each interaction point, both energy (\( \hbar \omega \)) and momentum (\( \hbar \mathbf{k} \)) are compelled to conserve in the cavity. This is called the “phase matching condition” in QO, which implies that the interaction always satisfies the resonance condition in the cavity, and is amplified maximally. As an example, if the laser beam with \( (\omega_L, \mathbf{k}_L) \) create \( n \) photons with energy and momentum, \( (\omega_1, \mathbf{k}_1), (\omega_1, \mathbf{k}_1), \cdots, (\omega_n, \mathbf{k}_n) \), then we have
\[
\omega_L = \omega_1 + \omega_2 + \cdots + \omega_n, \quad \text{and} \quad \mathbf{k}_L = \mathbf{k}_1 + \mathbf{k}_2 + \cdots + \mathbf{k}_n.
\]
It is important to note that in the processes occurring in the “ring cavity”, all the photons, including the laser photons, propagate in one-direction, and the motion in the opposite direction is prohibited.

The cavity called “optical parametric oscillator (OPO)” is an apparatus with one spacial dimension, such as a Fabry-Pérot resonator and a ring resonator. In Fabry-Pérot resonator, there are both forward and backward motions by mirror reflection, while in the ring resonator, the motion is restricted to the one-way round trip. Therefore, we will study this ring resonator in this paper, for simplicity. In such one-dimensional one-way motion, the energy-momentum conservation in Eq.(10) is reduced solely to the energy conservation,
\[
\omega_L = \omega_1 + \omega_2 + \cdots + \omega_n,
\]
where the momentum conservation is automatically guaranteed, since the magnitude of a momentum is proportional to its energy, \( |\mathbf{k}| = \omega/c' \).

The other possible processes are
\[
\omega_L + (\omega_1 + \omega_2 + \cdots + \omega_m) = \omega_{m+1} + \cdots + \omega_n,
\]
which implies that the laser beam and the photons with energy \( (\omega_1, \omega_2, \cdots, \omega_m) \) come into the cavity and annihilate, and the photons with \( (\omega_{m+1}, \cdots, \omega_n) \) are created and go out from the cavity.

Therefore, QO in one-dimensional apparatus can be analyzed without referring to the momentum or the space coordinate, if the apparatus is uniform in sufficiently long length scales in between mirrors.

Now, it is reasonable to study QO in terms of the following non-linear quantum mechanics with time-dependent interactions,
\[
L = \frac{1}{2} \dot{x}^2 - \frac{1}{2} \omega_0^2 x^2 - \frac{1}{2!} \lambda_2(t) x^2 - \frac{1}{4!} \lambda_4(t) x^4,
\]
We can include more higher-order terms such as \( x^6, x^8, \cdots \). Then, the quantum mechanics (QM) has non-linear interactions via the couplings \( \lambda_n(t) \), given in the interaction picture as
\[
L^{(n)} = -\frac{1}{n!} \lambda_n(t) \dot{x}(t)^n,
\]
where
\[
\hat{x}(t) = \sqrt{\frac{\hbar}{2\omega_0}} (\hat{a} e^{-i\omega_0 t} + \hat{a}^\dagger e^{i\omega_0 t})
\]
In comparison of Eq. (9) in QO with Eq. (14) in QM, we understand immediately that in case of one spacial dimension, these equations become equivalent under the following correspondence:

\[ \lambda_n(t) \leftrightarrow \varepsilon_0 \chi^{(n)} E_L(t), \quad \hat{x}(t) \leftrightarrow \hat{E}_q(t). \]  

The time dependent coupling \( \lambda_2(t) \) in QM and the non-linear dielectric constant \( \chi^{(2)} \) in QO equally generate the squeezing states, where the uncertainty for the “coordinate” is scaled up (down), while that of “momentum” is scaled down (up), keeping the product of them unchanged. In QO, the vector potential \( \mathbf{A}(t, x) \) and the electric field \( \mathbf{E}(t, x) \) form a canonical conjugate pair, playing the role of “coordinate” and “momentum”, respectively.

It is remarkable that the squeezing is observed in OPO, in which the Fabry-Pérot resonator is implemented with a non-linear crystal of MgO:LiNbO\(_3\) stored inside the cavity \([3]\).

The appearance of the squeezing states in the parametric down-conversion of laser photon \( \gamma_L \) into two degenerate photons \( \gamma_1 \) and \( \gamma_2 \) (\( \omega_L = \omega_1 + \omega_2 \) and \( \omega_1 = \omega_2 \)), is confirmed using the homodyne interferometer.

In this paper, we develop a formalism to study the phenomena a more generally “beyond the squeezing mechanism”, for example to see beyond the squeezing induced by the anharmonic coupling \( \lambda_4(t) \) in QM or by the non-linear dielectric constant \( \chi^{(4)} \) in QO. The more general case of beyond the squeezing, having the higher-order interactions, was studied separately in \([4]\), by referring to the Virasoro algebra, in which a general formula is derived, which gives the number of particles produced, under a condition of the number of particles produced is large.

Beyond the squeezing is also called “generalized squeezing” \([5]\). These generalized squeezed states are given by applying the following operator to the vacuum \( k \geq 3 \),

\[ \hat{U}_k = \exp \left( z \hat{a}^k + z^* \hat{a}^\dagger k \right) \]

which are naturally conceivable from the analogy of coherent states \( k = 1 \),

\[ \hat{U}_1 = \exp \left( z \hat{a} + z^* \hat{a}^\dagger \right), \]

and the squeezed states \( k = 2 \),

\[ \hat{U}_2 = \exp \left( z \hat{a}^2 + z^* \hat{a}^{\dagger 2} \right). \]

It is, however, controversial, whether the vacuum expectation value of the generalized squeezed state \( \langle 0 | \hat{U}_k^\dagger \hat{U}_k | 0 \rangle \) \( k \geq 3 \) is divergent or convergent. See Appendix for the detail.

In the next section, a ring resonator and its resonance condition are discussed. Transition amplitude for ring resonator is formulated in Section 3. In Section 4, the propagator and the vertex are explicitly given for QO inside ring resonator in the presence of NL-material, which are necessary items to estimate the transition amplitude. Transition amplitude is written using the propagators and the vertex functions in Section 5, so that our formalism can be applicable to higher-order calculations in QO in the cavity. The final section is devoted to conclusion and discussion; where possible application of the formalism developed in this paper is discussed, such as in estimating averaged number of produced photons, Husimi function and the observables to examine beyond the squeezing of photons. A controversial issue on generalized squeezing is given in Appendix.
2 Ring Resonator for Optical Parametric Oscillation

We adopt a “ring resonator” as the apparatus to observe the beyond squeezing (generalized squeezing) by Optical Parametric Oscillation (OPO), which is depicted in (Figure 1).

![Figure 1: A ring resonator as an apparatus to observe beyond the squeezing by Optical Parametric Oscillation (OPO).](image)

The ring resonator, confined in a cavity, consists of four mirrors, \(M_1 - M_4\). The \(M_1\) is the half-mirror, through which the input laser beam comes in from the left to the cavity, and \(M_2\) is the output half-mirror, through which the photons go out and are detected by the photo-diode (PD) located outside the cavity. The remaining \(M_3\) and \(M_4\) are ordinal mirrors. The non-linear (NL) crystal of length \(\ell\) is placed in between \(M_1\) and \(M_2\). The path length of light from \(M_1\) to \(M_2\) is \(L'\) and \(L\) is the length of a round trip in the resonator. The ring resonator extends in one spatial direction, along which the coordinate \(z\) is taken. Choosing \(z = 0\) in the center of the crystal, any path length is measured by the coordinate \(z\) from the origin, clock-wise along the direction of propagation of light.

The crystal with non-linear (NL) dielectric constants is assumed to have crystal planes perpendicular to \(z\). The planes are separated regularly by a separation of a lattice constant of the crystal, \(a_{\text{crys}}\).

We consider the laser beam to be an intense “classical wave” in this paper. This assumption is not bad, since above the threshold of laser oscillation (the pumping rate dominates over the loss in the cavity), laser photons form a coherent state, so that the laser light can be considered as a classical electric field \(E_L(t, z)\) with a well-defined phase without fluctuations \([1]\). The oscillation of this electric field of the laser in the perpendicular direction to \(z\), induces the oscillation of electrons on the crystal planes in the same perpendicular direction, from which photons are generated via the non-linear interaction in Eq. \([2]\).
First, we will estimate the resonance condition of the cavity. We denote $r_i$ the reflection coefficient of the i-th mirror, for the amplitude. Then, the corresponding transmission coefficient is $\sqrt{1 - r_i^2}$.

The electric field of light with angular frequency $\omega$ can be written as

$$E(t, z) = E e^{-i\omega(t - \frac{z}{c(z)}) - \gamma(z)z},$$

where the light velocity $c(z)$ can depend on the position $z$ to pass through; we choose $c$ in the vacuum and $c' = c/n_{crys}$ in the crystal with the refraction coefficient $n_{crys}$, while $\gamma(z)$ is the decay rate per unit length at $z$; we choose $\gamma(z) = 0$ in the vacuum and $\gamma(z) = \gamma'$ in the crystal.

Consider the light wave starting from $z$ at $t$. It undergoes round trips inside the cavity arbitrary times, being reflected by the mirrors, so that its electric field obtains an extra factor $K(\omega L/c)$, which includes both the phase change and the attenuation:

$$K(\omega L/c) = \frac{1}{1 - (r_1r_2r_3r_4)e^{i\phi(\omega L/c) - \gamma'\ell}},$$

where

$$\phi(\omega L/c) = \frac{\omega}{c}(L - \ell) + \frac{\omega}{c'}\ell.$$

So, the strength of the electric field is proportional to $|K|^2$, which gives a resonance behavior:

$$|K(\omega L/c)|^2 = \frac{1}{\{1 - re^{-\gamma'\ell}\}^2 + 4re^{-\gamma'\ell}\sin^2\{\frac{1}{2}\phi(\omega L/c)\}},$$

where $r = r_1r_2r_3r_4$.

If the resonance point at which $|K|^2$ takes the maximum value, is denoted by $\omega_R$ or its frequency $\nu_R$, we have the resonance condition as follows:

$$\frac{\phi(\omega_R L/c)}{2\pi} = \left(\frac{\omega_R}{\omega_{FSR}}\right) \left(1 + \frac{\ell}{L}(n_{crys} - 1)\right) = \text{integer},$$

where $\omega_{FSR} = 2\pi c/L$ is the angular frequency of a fictitious wave with wave length $L$, which is called the “free streaming range”, since adjacent resonances are separated by $\omega_{FSR}$. The full width at half maximum $(\omega_R/\omega_{FSR})_{FWHM}$ of the resonance is roughly

$$\left(\frac{\omega_R}{\omega_{FSR}}\right)_{FWHM} \approx \frac{|1 - re^{-\gamma'\ell}|}{\sqrt{re^{-\gamma'\ell}}} \ll 1,$$

since $r \approx 1$.

Therefore, we can tune $L$ very precisely so that the waves resonate and are enhanced in the cavity. The resonance is very sharp, and $\omega_R$ can be precisely selected. It is important to note here that if $\omega_1$ satisfies the minimum resonance condition:

$$\frac{\phi(\omega_1 L/c)}{2\pi} = \left(\frac{\omega_1}{\omega_{FSR}}\right) \left(1 + \frac{\ell}{L}(n_{crys} - 1)\right) = 1,$$
then, $2\omega_1, 3\omega_1, 4\omega_1, \cdots$ also satisfy the resonance condition Eq.(24) by choosing the integer as $2, 3, 4, \cdots$, if the refraction index $n_{\text{crys}}$, (generally depends on the frequency $\omega$), is constant for these waves. The FWHM in Eq.(25) depends on the waves, since it depends on the $r$ and $\gamma'$ whose dependency on $\omega$ may not be ignored.

We consider a crystal, having NL dielectric constants $\chi^{(2)}$ and $\chi^{(4)}$. Generalization to more general cases with $\chi^{(i)}$ ($i = 3, 5, 6, \cdots$) is also possible.

Now, we choose the laser frequency to be $\omega_L = \omega_0$, then the down-converted photons with frequencies, $\omega_L/4 = \omega_1$ and $\omega_L/2 = \omega_2 = 2\omega_1$, can also resonate and survive in the cavity.

In the cavity, the momenta of photons should always be parallel to the longitudinal direction. That is, if the photon in the cavity is scattered by the crystal and loses energy or alters the direction of momentum, then this photon can not exist in the cavity and disappears; the damping rate $\gamma'$ comes from these scatterings.

3 Transition amplitude for Ring Resonator

In this section, we will study the transition amplitude in QO, which describes the creation and annihilation of photons from a non-linear crystal, induced by the laser. In this QO inside the resonator, the Hamiltonian can be divided into two parts; the first part is denoted as $H_0$, which is time dependent and not free but is “quasi-free”, including the effect of damping in the cavity and the reflection and penetration by the mirrors. The second part $H_1$ gives the real interaction (creation and annihilation) of photons by the non-linear crystal. Thus, the Hamiltonian which we are going to study is time dependent, even in the “Shrödinger picture”:

$$ \hat{H}(t) = \hat{H}_0(t) + \hat{H}_1(t). $$

(27)

We will call $\hat{H}_0$ the “quasi-free Hamiltonian”, and $\hat{H}_1$ the “real interaction Hamiltonian”.

We will mimic the process of picture changing from the “Schrödinger picture” to the “interaction picture”, by treating $\hat{H}_0$ as if it was the free Hamiltonian.

If we introduce $\hat{U}_0(t)$, $U$-matrix or the time evolution operator for the quasi-free Hamiltonian $\hat{H}_0(t)$, it satisfies

$$ i\hbar \frac{\partial}{\partial t} \hat{U}_0(t) = \hat{H}_0(t)\hat{U}_0(t). $$

(28)

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2The ring resonator which we are considering is especially suited for observing the parity violation effects from $\chi^{(i)}$, $i =$odd integer. See for example [7], in which the ring resonator is examined to see the parity violation effect of neutrinos as dark matter, without QED background. On the other hand, the forward and backward motions in Fabry-Pérot cavity wash out the parity violation effects.

3To input the laser beam smoothly into the ring cavity and to extract the output photons effectively from it, some devise may be necessary. Referring to the gravitational wave detector [8], a way is to prepare two more cavities, the laser input cavity (LIC) and the signal extraction cavity (SEC). The former cavity (LIC) consists of the mirror $M_1$ and mirrors in front and behind of $M_1$ along the direction of laser beam, while the latter cavity (SEC) consists of the mirror $M_2$ and mirrors in front and behind of $M_2$ along the path to the photo-detector. The LIC can be designed almost transparent for $\omega_L$ but very reflective for $\omega_1$ and $\omega_2$, while the SEC can be designed very reflective for $\omega_L$ but almost transparent for $\omega_1$ and/or $\omega_2$. If necessary, the advanced technique such as modulation and demodulation of amplitude (AM) and of phase (PM) can be used.
Explicitly, we have
\[ \dot{U}_0(t) = T e^{\int_0^t dt' \hat{H}_0(t') / i\hbar}. \] (29)

In this “Schrödinger picture”, the equations of motion of wave function \( \psi(t)_S \) and operator \( \hat{O}_S \) are given by
\[ i\hbar \frac{\partial}{\partial t} \psi(t)_S = \hat{H}(t)\psi(t)_S, \quad \text{and} \quad i\hbar \frac{\partial}{\partial t} \hat{O}_S = 0, \] (30)

After moving to the “interaction picture” we have
\[ i\hbar \frac{\partial}{\partial t} \hat{O}_I = [\hat{O}_I, \hat{H}_0(t)_S], \quad \text{and} \quad i\hbar \frac{\partial}{\partial t} \psi(t)_I = \hat{H}_I(t) \psi(t)_I, \] (31)

where
\[ \psi(t)_I = \hat{U}_0^{-1}(t) \psi(t)_S, \quad \text{and} \quad \hat{H}_I(t)_I = \hat{U}_0(t)^{-1} \hat{H}_I(t) \hat{U}_0(t). \] (32)

Then, in the time evolution of the “interaction picture”, the quasi-free motion is subtracted from that of the “Schrödinger picture”. The motion of laser light and photons, before and after the real interaction in the cavity occurs, is quasi-free, so that in these periods the wave function in the “interaction picture” does not evolve.

The transition amplitude of QO in the ring resonator can be expressed iteratively as usual, by solving the time evolution of wave function in the “interaction picture”. We will use the number representation, in which the state is expanded in \( |n\rangle \) having \( n \) photons. Then, the Schrödinger equation in the interaction picture, \( i\hbar \frac{\partial}{\partial t} |n, t\rangle = \hat{H}_I(t)|n, t\rangle \), can be solved, if we choose the initial condition \( |n, -\infty\rangle = |m\rangle \), as follows

\[ |n, t\rangle = |m\rangle + \frac{1}{i\hbar} \int_{-\infty}^t dt_1 |n\rangle \langle n| \hat{H}_I(t_1)|m\rangle \]
\[ + \left( \frac{1}{i\hbar} \right)^2 \int_{-\infty}^t dt_1 \int_{t_1}^t dt_2 |n\rangle \langle n| \hat{H}_I(t_1)|m_1\rangle \langle m_1| \hat{H}_I(t_2)|m\rangle + \cdots \] (33)
\[ = |m\rangle + \frac{1}{i\hbar} \int_{-\infty}^t dt_1 |n\rangle \langle n| \hat{U}_0(t)^{-1}|n'\rangle \]
\[ \times \langle n'| \hat{U}_0(t_1, t_1)|m_1\rangle \langle m_1| \hat{H}_I(t_1)|m_2\rangle \langle m_2| \hat{U}_0(t_1, -\infty)|m'\rangle \times \langle m'| \hat{U}_0(-\infty)|m\rangle \]
\[ + \left( \frac{1}{i\hbar} \right)^2 \int_{-\infty}^t dt_1 \int_{t_1}^t dt_2 |n\rangle \langle n| \hat{U}_0(t)^{-1}|n'\rangle \]
\[ \times \langle n'| \hat{U}_0(t_1, t_1)|m_1\rangle \langle m_1| \hat{H}_I(t_1)|m_2\rangle \langle m_2| \hat{U}_0(t_1, t_2)|m_3\rangle \langle m_3| \hat{H}_I(t_2)|m_4\rangle \langle m_4| \hat{U}_0(t_2, -\infty)|m'\rangle \]
\[ \times \langle m'| \hat{U}_0(-\infty)|m\rangle + \cdots, \] (34)

where \( \hat{U}_0(t_1, t_2) \equiv \hat{U}_0(t_1) \hat{U}_0(t_2)^{-1} \) is a transition amplitude from \( t_2 \) to \( t_1 \) by the “quasi-free Hamiltonian \( \hat{H}_I \)”. When \( t \) is finite, then the factor \( \langle n| \hat{U}_0(t)^{-1}|n'\rangle \) should be carefully estimated, but in our case, we will take \( t = -\infty \), and consider the transition amplitude from \( t = -\infty \) to \( t = \infty \), that is the S-matrix, we may choose
\[ \langle n| \hat{U}_0(\infty)^{-1}|n'\rangle = \delta_{nn'}, \quad \langle m'| \hat{U}_0(-\infty)|m\rangle = \delta_{mm'}. \] (35)
Next, we have to discuss an important point that QO is essentially a field theory, having spacial degrees of freedom, \(x\). Then, the Hamiltonian is given as the integral over \(x\) of the “Hamiltonian density \(H(t, x)\)”, that is
\[
\hat{H}_0(t) = \int dx \, \mathcal{H}_0(t, x), \quad \text{and} \quad \hat{H}_1(t) = \int dx \, \mathcal{H}_1(t, x).
\]

Thus, the transition amplitude in QO can be written as follows:
\[
\langle n, t \rangle = \langle m \rangle + \frac{1}{\sqrt{i\hbar}} \int_{-\infty}^t dt_1 \int dx_1 \langle n | \hat{U}_0(t)^{-1} | n' \rangle \\
\times \langle n' | \hat{U}_0(t_1) | m_1 \rangle \langle m_1 | \mathcal{H}_1(t_1, x_1) | m_2 \rangle \langle m_2 | \hat{U}_0(t_1, -\infty) | m' \rangle \\
\times \langle m' | \hat{U}_0(-\infty) | m \rangle \\
+ \left( \frac{1}{\sqrt{i\hbar}} \right)^2 \int_{-\infty}^t dt_1 \int dx_1 \int_{-\infty}^{t_1} dt_2 \int dx_2 \langle n | \hat{U}_0(t)^{-1} | n' \rangle \\
\times \langle n' | \hat{U}_0(t_{11}) | m_1 \rangle \langle m_1 | \mathcal{H}_1(t_{11}, x_1) | m_2 \rangle \langle m_2 | \hat{U}_0(t_1, t_2) | m_3 \rangle \\
\times \langle m_3 | \mathcal{H}_1(t_2, x_2) | m_4 \rangle \langle m_4 | \hat{U}_0(t_2, -\infty) | m' \rangle \times \langle m' | \hat{U}_0(-\infty) | m \rangle \\
+ \cdots.
\]

\[37\]

4 Propagator and Vertex inside Ring Resonator containing Non-linear Material

We first examine a simple case of quantum optics (QO) inside the uniformly distributed non-linear material, which was discussed a bit in Introduction. The Lagrangian density in this case is Eq.(4),
\[
\mathcal{L}_{QO} = \frac{1}{2} (E \cdot D - H \cdot B) = \mathcal{L}_0 + \mathcal{L}_1,
\]
where
\[
\mathcal{L}_0 = \frac{1}{2} \varepsilon \left( E^2 - \frac{1}{\varepsilon \mu_0} B^2 \right), \quad \text{and} \quad \mathcal{L}_1 = \frac{1}{2} \varepsilon_0 \sum_{n \geq 2} \chi^{(n)}(E)^{n+1}.
\]

The quadratic terms are combined into a quasi-free Lagrangian with a new parameter \( \varepsilon = \varepsilon_0 (1 + \chi^{(1)}) \), the dielectric constant of the material.

In terms of vector potential \( \mathbf{A}(t, \mathbf{x}) \) and electric field \( \mathbf{E} = -\dot{\mathbf{A}} \), the conjugate momentum of the former is given by \( \Pi_A = -\varepsilon E = \varepsilon \dot{A} \), and \( \mathbf{B} = \nabla \times \mathbf{A} \), so that the Hamiltonian density \( \mathcal{H} \) reads
\[
\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1,
\]
where
\[
\mathcal{H}_0 = \frac{1}{2} \varepsilon \left( E^2 + \frac{1}{\varepsilon \mu_0} B^2 \right), \quad \text{and} \quad \mathcal{H}_1 = \frac{1}{2} \varepsilon_0 \sum_{n \geq 2} n \chi^{(n)}(E)^{n+1}.
\]

The quasi-free Hamiltonian, in this case, is a free Hamiltonian in the medium, where \( c' = c/n = c/\sqrt{\varepsilon \mu_0} \) (\( n \): the refraction constant) is the light velocity in the medium.

Now, we are ready to study QO inside the ring resonator.
4.1 Mode Expansion

In QED, any mode having an arbitrary wave vector \( \mathbf{k} \) have to be considered, but in QO, only a few modes, having specific values of wave vectors, have to be considered due to the resonance condition of the cavity. In this situation, it is better to formulate QO in terms of discretized coordinates and momenta. Let us denote discretized coordinates with equal spacing \( a \) by \( \mathbf{x}_n \), and the discretized (angular frequency \( \omega \), wave vector \( \mathbf{k} \)) with equal spacing by \( (\omega_m, \mathbf{k}_m) \). The total number of them are equally \( N(\gg 1) \), that is, \((n, m) = 1, 2, \cdots, N,\) and \(Na = L \) is the total length of the system. Then, the spacing of the wave vector is \( 2\pi/L \) as usual. Corresponding to the mode expansion of \( \mathbf{E} \) in Eq. [3], the mode expansion of the vector potential \( \mathbf{A} \) and its canonical conjugate \( \Pi_A \), are as follows:\[4\]

\[
\hat{A}_q(t, \mathbf{x}) = \int d^3k \sqrt{\frac{\hbar}{(2\pi)^3 \varepsilon 2\omega}} (\hat{a}(\mathbf{k}) e^{-i(\omega t - \mathbf{k} \cdot \mathbf{x})} + \hat{a}^\dagger(\mathbf{k}) e^{i(\omega t - \mathbf{k} \cdot \mathbf{x})}), \tag{42}
\]

\[
\hat{\Pi}_{A,q}(t, \mathbf{x}) = \varepsilon \int d^3k \sqrt{\frac{\hbar}{(2\pi)^3 \varepsilon 2\omega}} (-i\omega) (\hat{a}(\mathbf{k}) e^{-i(\omega t - \mathbf{k} \cdot \mathbf{x})} - \hat{a}^\dagger(\mathbf{k}) e^{i(\omega t - \mathbf{k} \cdot \mathbf{x})}), \tag{43}
\]


\[
\left[ \hat{A}_q(t, \mathbf{x}), \hat{\Pi}_{A,q}(t, \mathbf{x}') \right] = i\hbar \delta^{(3)}(\mathbf{x} - \mathbf{x'}), \quad \text{or equivalently,} \quad \left[ \hat{a}(\mathbf{k}), \hat{a}^\dagger(\mathbf{k'}) \right] = \delta^{(3)}(\mathbf{k} - \mathbf{k'}) . \tag{44}
\]

If we restrict the problem to QO in one-dimensional cavity, the discretized version of operators can be introduced by\[5\]

\[
\sqrt{a} \hat{A}_q(t, \mathbf{x}) = \hat{A}(t, \mathbf{x}_n), \quad \sqrt{a} \hat{\Pi}_{A,q}(t, \mathbf{x}) = \hat{\Pi}_{A,q}(t, \mathbf{x}_n), \tag{45}
\]

\[
\sqrt\frac{2\pi}{L} \hat{a}(\mathbf{k}) = \hat{a}(\mathbf{k}_m), \quad \sqrt\frac{2\pi}{L} \hat{a}^\dagger(\mathbf{k}) = \hat{a}^\dagger(\mathbf{k}_m). \tag{46}
\]

Then, the commutation relations are expressed with Kroneker’s delta as follows:

\[
\left[ \hat{A}_q(t, \mathbf{x}_n), \hat{\Pi}_{A,q}(t, \mathbf{x}_{n'}) \right] = i\hbar \delta_{nn'}, \quad \text{or equivalently,} \quad \left[ \hat{a}(\mathbf{k}_m), \hat{a}(\mathbf{k}_{m'}) \right] = \delta_{mm'} . \tag{47}
\]

Thus, the discretized version of the vector potential and the electric field are given by\[6\]

\[
\hat{A}_q(t, z_n) = \sum_{m=1}^{N} \sqrt{\frac{\hbar}{N \varepsilon 2\omega_m}} (\hat{a}_m e^{-i(\omega_m t - \mathbf{k}_m z_n)} + \hat{a}_m^\dagger e^{i(\omega_m t - \mathbf{k}_m z_n)}), \tag{48}
\]

\[
\hat{E}_q(t, z_n) = \sum_{m=1}^{N} \sqrt{\frac{\hbar}{N \varepsilon 2\omega_m}} (i\omega_m) (\hat{a}_m e^{-i(\omega_m t - \mathbf{k}_m z_n)} - \hat{a}_m^\dagger e^{i(\omega_m t - \mathbf{k}_m z_n)}). \tag{49}
\]

These equations are valid for any spacial dimension, if we consider \( N \) to be the total number of discretized points in coordinate space and wave vector space. Similarly, the classical part of laser in Eq. [3], is renormalized, when using the discrete coordinates,

\[
\sqrt{a} E_L(t, z) = E_L(t, z_n) \text{ with } \left\{ \begin{array}{ll} E_L(t, z) = E_L e^{-i\omega_L(t-z/c(z))} + E_L^\dagger e^{i\omega_L(t-z/c(z))}, \\
E_L(t, z_n) = E_L e^{-i\omega_L(t-z_n/c(z))} + E_L^\dagger e^{i\omega_L(t-z_n/c(z))}. \end{array} \right. \tag{50}
\]

\[4\] The reason why the dielectric constant \( \varepsilon \) appears in the mode expansion is that the conjugate momentum defined by \( \Pi_A = \varepsilon \hat{q} A \) in the medium.

\[5\] In case of three spacial dimensions, the square roots should be replaced by \( a^{\frac{2}{3}} \) or \( (\frac{2\pi}{T})^\frac{2}{3} \).
In the above, the discrete sum over coordinates and that over wave vectors appear. These two discretizations are correlated, but it is convenient to convert, while keeping the discrete sum over modes, the discrete sum to the coordinate integration, by using \( \sum_n = \int dz/a \). This is valid since \( a \) is small compared to the wave length. The discretization of modes appears not from the correspondence between \( a \) and \( L \), but from the resonance condition. In the following, we adopt this hybrid expression, that is, coordinate is continuous, while the wave vector is discrete.

In our ring cavity, only two specific modes of photons, having angular frequencies, \( \omega_1 = \omega_L/4 \) and \( \omega_2 = \omega_L/2 \) are relevant.

Therefore, in the hybrid expression, the classical part of electric field from laser is the same as Eq.(8), but the quantum part of the electric field is chosen as follows, namely

\[
\hat{E}_q(t,z) = i \sum_{i=1,2} \sqrt{\frac{\hbar \omega_i}{2L \varepsilon(z)}} \left( \hat{a}_i e^{-i(\omega_i(t-z/c(z))} - \hat{a}_i^\dagger e^{i\omega_i(t-z/c(z))} \right),
\]

where the system size \( L \) remains, and \( c(z), \varepsilon(z) \) are the light velocity and the dielectric constant at a position \( z \), respectively. It is natural to choose \( L \) be a total path length of the ring resonator.

### 4.2 Propagator

The “propagator” in the number representation is given as

\[
\langle n|\hat{U}_0(t,t')|n'\rangle = \langle n|\hat{U}_0(t)\hat{U}_0(t')^{-1}|n'\rangle.
\]

Here, \( \hat{U}(t,t') \) satisfies Eq.(51), so that it is true to give the phase change of wave in time, \( e^{-\frac{\hbar}{\sqrt{2L}}(t-t')} = (e^{-\frac{i\omega(t-t')}{c}})^n \) for the \( n \) photon state, but that is not all. The propagation of laser beam and of produced photons move clock-wisely inside the cavity, inside which non-linear material and the mirrors exist, so that the wave changes position dependently. In our case with one spacial dimension, the position \( z \) is determined by the time \( t \). Therefore, we will write the propagator as a time-ordered product, being defined piece-wisely. As an example, let us consider a photon with the frequency \( \omega \), at a position \( z \) in the vacuum, and finally arrives at \((t_3, z_3)\). The corresponding propagator can be

\[
U(t_3, t_0) = U(t_3, t_2, \text{vac}) \times r \times U(t_2, t_1; \text{vac}) \times U(t_1, t_0; \text{medium}),
\]

where \( U(t_1, t_0; \text{vac}) = e^{-i\omega(t_1-t_0)-(z_1-z_0)/c} \), but in the medium, damping occurs with rate \( \gamma' \) per length, so that \( U(t_1, t_0; \text{medium}) = e^{-i\omega(t_1-t_0)-(z_1-z_0)/c'-\gamma'(z_1-z_0)} \). The derivation of the propagator is the same both in the classical wave and the quantum wave.\(^6\)

\(^6\)If we wish to derive more rigorously the propagators, we can do it, by considering the Hilbert space of photons as a product of that inside the cavity (or that of the system) \( \mathcal{H}_s \) and that outside the cavity (or that of the heat bath) \( \mathcal{H}_b \), namely, \( \mathcal{H} = \mathcal{H}_s \otimes \mathcal{H}_b \). Since the attenuation caused by \( \gamma' \) and \( r \) is the effect of escape of photons from the system to the heat bath. If we trace out the heat bath and concentrate to the system, we have the effect of attenuation, caused by the crystal, and the reflection and penetration via mirrors. The rigorous result does not, however, differ from that given in the above.
Now, we can understand that the propagator of \( n \) photons is
\[
\langle n | \hat{U}_0(t, t') | n' \rangle \equiv \delta_{nn'} D_n(t, z; t', z'), \quad \text{and} \quad D_n(t, z; t', z') = \langle 1 | \hat{U}_0(t, t') | 1 \rangle^n \equiv D(t, z; t', z')^n,
\]
where \( 1/n! \) appeared from the normalization of the state is cancelled by the degeneracy \( n! \) which comes from the ways of connecting \( n \) (initial and final) photons by propagators.

The propagator of a single photon, \( D(t, z; t', z') \), describes the transition amplitude of state by the quasi-free Hamiltonian. Its derivation is the same as that of the coefficient \( K(\omega L/c) \) in Eq.\((2)\), including the round trips. Difference between \( K \) and the propagator \( D \) is that the starting point \( (t', z') \) and the arriving point \( (t', z') \) are equal in the former, but different in the latter.

There are a number of propagators:
1) “propagator” from a crystal plane \( C' \) to another crystal plane \( C' \):
\[
D_{C,C'}^\omega(t, z; t', z') = \frac{e^{-i\omega(t-t') - (z-z')/c'}}{1 - (r_\omega e^{i\phi(\omega L/c)} - \gamma')} \equiv e^{-i\omega((t-t') - (z-z')/c')} K_{C,C'}^\omega(z, z').
\]
Here, \( \phi(\omega L/c) \) is given in Eq.\((22)\) in the previous section. This is applicable to \( \omega = \omega_1 \), or \( \omega_2 (= 2\omega_1) \). If the \( n \) photons propagate, then the factor becomes \( (D_{C,C'}^\omega(t, z; t', z'))^n \).

2) “propagator” for the laser light:
The \( \hat{U}_0(t, t') \) is purely quantum object, giving the transformation of operators from the Shrödinger picture to the interaction picture. Therefore, there is no such concept of “propagator” for the purely classical laser beam. However, it is reasonable to use the name “propagator”, \( \hat{U}(t, t') \) also for the classical electromagnetic field, since the time evolution factor is the same as in the quantum theory. Then, the propagator of the classical laser beam is
\[
D_{C,C'}^L(t, z; t', z') = \frac{e^{-i\omega_L((t-t') - (z-z')/c')}}{1 - r_L (e^{i\phi(\omega L/c)} - \gamma')} \equiv e^{-i\omega_L((t-t') - (z-z')/c')} K_{C,C'}^L(z, z').
\]
Here, we have ignored the damping effect of \( \gamma' \), since it should be cancelled by the strong pumping of the laser beam.

3) “propagator” from the final interaction point to the outside photo-detector (PD):
This “propagator” has an additional effect to the propagator, which is caused by passing through the mirror \( M_2 \), and by traveling from the final interaction point \( z_f \) to the photo-detector located outside at a distance \( z - L'/2 \) from the mirror \( M_2 \).

We denote the final interaction plane be \( C_f \), then we have
\[
D_{PD,C_f}^\omega(t, z; t_f, z_f) = \frac{\sqrt{1 - r_f^2} e^{-i\omega((t-t_f)-(L'/2-z_f)/c') - (z-L'/2)/c'} - \gamma'(e^{i\phi(\omega L/c)} - \gamma'):C_f \rangle \equiv e^{-i\omega((t-t_f)-(L'/2-z_f)/c') - (z-L'/2)/c'} K_{PD,C_f}^\omega(z, z_f).
\]
4.3 Vertex

The real interaction of our problem occurs at the crystal planes separated by $a_{\text{crys}}$, so that the “vertex” is given in the vicinity of the crystal plane, giving

$$\langle m | \hat{\mathcal{H}}_1(t) | m' \rangle = \int_{z-a_{\text{crys}}/2}^{z+a_{\text{crys}}/2} dz' \langle m | \hat{\mathcal{H}}_1(t,z') | m' \rangle,$$

(59)

where we have assumed the local interaction at $z$. In the above expression we put the suffix $S$ as $\hat{\mathcal{H}}_1(t,z)_S$, to clarify the “Shrödinger picture”. We assume as usual that the operators in the “Shrödinger picture” and the “interaction picture” coincide at $t = 0$.

We will write the interaction Hamiltonian, after separating the dominant classical part of laser from the quantum part of photons, as follows:

$$\langle m | \hat{\mathcal{H}}_1(t)_S | m' \rangle = \int_{z-a_{\text{crys}}/2}^{z+a_{\text{crys}}/2} dz' \langle m | \hat{\mathcal{H}}_1(t,z')_S | m' \rangle,$$

(60)

$$= \frac{1}{2} \varepsilon_0 \sum_{n \geq 2} n(n+1) \chi^{(n)} \int_{z-a_{\text{crys}}/2}^{z+a_{\text{crys}}/2} dz' E_L(t,z') \times \langle m | (\hat{E}_q(t = 0,z'))^n | m' \rangle.$$

(61)

In the following, we study a simple NL case of having only $\chi^{(2)}$ and $\chi^{(4)}$, and hence the real interaction Hamiltonian takes the following form:

$$\hat{\mathcal{H}}_1(t,z)_S = \frac{1}{2} \varepsilon_0 E_L(t,z) \times \left\{ 6 \chi^{(2)} (\hat{E}_q(0,z))^2 + 20 \chi^{(4)} (\hat{E}_q(0,z))^4 \right\}.$$

(62)

The real interaction occurs inside the non-linear crystal, where the light velocity is $c'$, so that the mode expansion used in writing the vertex function is

$$\left\{ \begin{array}{ll}
\text{(classical part)} & : E_L(z)_S = E_L e^{i\omega_L z/c'} + E_L^\dagger e^{-i\omega_L z/c'}, \\
\text{(quantum part)} & : \hat{E}_q(z)_S = i \sum_{i=1,2} \sqrt{\frac{\hbar \omega_i}{2 E_L}} \left( \hat{a}_i e^{i\omega_i z/c'} - \hat{a}_i^\dagger e^{-i\omega_i z/c'} \right),
\end{array} \right.$$

(63)

where all the classical and quantum photons are assumed to propagate in the positive (clockwise) direction along the cavity, and the frequency is restricted to $\omega_1 = \omega_L/4$ and $\omega_2 = \omega_L/2$, by the resonance condition of the cavity.

This is a good place to discuss the momentum conservation. From the position integral near the vertex point, if the space near the vertex point is uniform (i.e. the relevant wavelength $\lambda$ of laser beam and photons satisfies $\lambda \ll a_{\text{crys}}$, then the momentum conservation holds, and the sum of all the wave vectors coming in the vertex is equal to the sum of all the wave vectors coming out from the vertex. This property also holds in the usual old fashioned perturbation theory.

As for the energy conservation, it does not hold in the old fashioned perturbation theory, since the transition amplitude becomes singular, if the energy conservation at each interaction point holds. This problem is solved in this paper, by averaging the frequency of laser beam around the resonance point. (See the details in the next section.)

\footnote{We do put $S$ for the classical laser beam as well as the quantum photons, in order to indicate that all the time dependencies have been absorbed into the classical and quantum propagators.}
Now, the possible interactions can be listed as follows:

\[ \chi^{(4)} = \begin{cases} 
\{4\}_4 & : \text{Laser emits 4}\gamma_1 s \text{ at a crystal surface, } (\omega_L = 4\omega_1 \to 4\omega_1), \\
\{4\}_-4 & : \text{4}\gamma_1 s \text{ are absorbed by Laser at the surface, } (4\omega_1 \to \omega_L = 4\omega_1), \\
\{4\}_2 & : \text{Laser + 1}\gamma_1 \text{ emitts } 2\gamma_2 s + 1\gamma_1 \text{ at a crystal surface, } \\
& \quad (\omega_L + \omega_1 \to \omega_1 + 2\omega_2), \\
\{4\}_-2 & : 2\gamma_2 s + 1\gamma_1 \text{ emit Laser + 1}\gamma_1 \text{ at a crystal surface, } \\
& \quad (2\omega_2 + \omega_1 \to \omega_L + \omega_1). 
\end{cases} \]

\[ \chi^{(2)} = \begin{cases} 
\{2\}_2 & : \text{Laser emits } 2\gamma_2 s \text{ at a crystal surface, } (\omega_L = 2\omega_2 \to 2\omega_2), \\
\{2\}_-2 & : 2\gamma_2 s \text{ are absorbed by Laser at the surface, } (2\omega_2 \to 2\omega_2 = \omega_L). 
\end{cases} \]

where we denote the photons with energy \( \omega_1 \) and \( \omega_2 \) as \( \gamma_1 \) and \( \gamma_2 \), respectively. The different types of interactions are labeled by \( \{4\}_\pm, \{4\}_\pm, \text{ and } \{2\}_\pm \) to make the individual processes clearer. It is noted that the other processes, such as \( (\omega_L + 1\gamma_1 \leftrightarrow 3\gamma_1) \) does not occur, since the energy conservation is violated.

5 General Formula of Transition Amplitude or S-matrix in QO inside Cavity

To obtain the general formula of transition amplitude or S-matrix, we first perform integrations over times \( (t_1, t_2, t_3, \cdots) \) of the product of vertex operators, which are arranged according to the time ordering.

Time dependence of the vertex is \( E_{\ell} e^{-i\omega_{\ell} t} \) or \( E_{\ell}' e^{i\omega_{\ell} t} \), and that of propagator \( D(t - t') \) is \( e^{\pm i\omega(t-t')} \), and hence the time integration is the same as in quantum mechanics. Here, we write the vertex with “energy” eigen-states as \( \langle E_k | V(t_k) | E_l \rangle \), where \( E_k \) and \( E_l \) are energies before and after the k-th interaction. Here we do not distinguish the “energy” and the “angular frequency”.

Then, up to position dependent extra factors, \( K_s \), the transition amplitude \( S(t, -\infty) \), defined by \( \psi(t) = S(t, -\infty) \psi(-\infty) \), reads perturbatively,

\[
S(t, -\infty)|_{\text{up to } K_s} = 1 + \sum_{N=1}^{\infty} \left( \frac{1}{\hbar} \right)^N \int_{-\infty}^{t_1} \int_{-\infty}^{t_2} \cdots \int_{-\infty}^{t_N} e^{-i(\omega_1 t + E_i - E_f + i\epsilon) t_N} \langle E_f | \hat{H}_1 | E_1 \rangle \\
\times \langle E_{k_2} | \hat{H}_1 | E_2 \rangle \times \cdots \times e^{-i(\omega_{k_2} + E_{\ell} - E_{k_2} + i\epsilon) t_N} \langle E_{k_N} | \hat{H}_1 | E_{N} \rangle \\
= 1 + \sum_{N=1}^{\infty} e^{-i(\omega_1 t + \cdots + \omega_{k_N} + E_i - E_f)} \prod_{k=1}^{N} \langle E_k | \hat{H}_1 | E_k \rangle / \hbar \square \left( \omega_k + \cdots + \omega_{k_N} \right) + E_i - E_f + i\epsilon,
\]

(67)
where \( E_i = E_N \) and \( E_f = E'_1 \) are initial and final “energies” of photons, respectively. The “energies” before and after each interaction can be expressed by using the number representation as \( \langle (n_k)_1, (n_k)_2 | H_1(t_k) | (n_k)_1 - (s_k)_1, (n_k)_2 - (s_k)_2 \rangle \), where the suffix 1 and 2 indicate the particle numbers \( (n_k)_1,2 \) and their difference \( (s_k)_1,2 \) be for the two kinds of photons, \( \gamma_1 \) and \( \gamma_2 \), respectively. Then, we have

\[
E'_1 = E_i + (s_k + s_{k+1} + \cdots + s_N)_1 \omega_1 + (s_k + s_{k+1} + \cdots + s_N)_2 \omega_2, 
\]

where \( \omega_1 \) and \( \omega_2 \) are frequencies of photon \( \gamma_1 \) and \( \gamma_2 \), respectively.

The formula obtained as Eq.(67) is nothing but the formula of old fashioned perturbation theory in QM. When we apply it to our QO, the phase matching condition tells that the laser photon has a definite wave vector \( \omega \) at each interaction point \( k \). This averaging is a kind of tuning of the laser beam at the resonance point.

A candidate of the weight for this averaging is a Gaussian distribution, \( W(|\omega_L|) \), with a center \( \omega_R \) and a width \( \delta \).

\[
W(|\omega_L^k|) = \frac{1}{\sqrt{2\pi} \delta} e^{-((\omega_L^k-\omega_R)^2)/2\delta^2}. 
\]

This averaging implies that the laser photon has a definite wave vector \( k^L = \pm \omega_R/c' \) at the interaction plane of crystal, but its energy is not exactly \( \pm \omega_R \), but is distributed as

\[
\omega_R - \delta < |\omega_L^k| < \omega_R + \delta. 
\]

Then, the averaging over the laser frequency becomes

\[
\langle S(t, -\infty) |_{\text{up to } Ks} \rangle_{\omega_L} = 1 + \sum_{N=1}^{\infty} \prod_{k=1}^{N} \int_{-\infty}^{\infty} d \omega_L^k W(|\omega_L^k|) S(t, -\infty; \{\omega_L^k\}) |_{\text{up to } Ks}. 
\]

Since the weight function vanishes at infinitely large frequency,

\[
W(|\omega_L^k|) \rightarrow 0, \text{ for } |\omega_L^k| \rightarrow \infty. 
\]

we can perform the contour integration clock-wisely by the new variables with tilde, defined by

\[
\tilde{\omega}_1^L = \omega_1^L + \cdots \omega_N^L, \quad \tilde{\omega}_2^L = \omega_2^L + \cdots \omega_N^L, \ldots, \quad \tilde{\omega}_N^L = \omega_N^L. 
\]

\(^6\)Corresponding to absorption and emission of laser light, we consider \( \omega^L \) as positive and negative values, respectively.

\(^8\)At first sight, this averaging seems to violate the “massless property” of photon, but is a standard way to tame the mass singularity. Physically, this \( \delta \) can be the ambiguity to detect photon energy, which is inevitable. In QO, it can be the “ambiguity of tuning the laser frequency at the resonance point”. It can also be that the laser beam is not a plane wave but the wave packet.
and pickup the pole residues. The choice of pole residues recovers the energy conservation at each vertex, \( \omega_k^L = E'_k - E_k \), but \( \omega_k^L \) is not definitely fixed, but is distributed with a sharp peak around \( \pm \omega_R \). Then, we arrive at

\[
\langle S(t, -\infty) \rangle_{\text{up to } K_s} = 1 + \sum_{N=1}^{\infty} \prod_{k=1}^{N} \left( -2\pi i / \hbar \right) W(|E'_k - E_k|) \langle E'_k | \hat{H}_1 | E_k \rangle, \tag{74}
\]

where

\[
W(|E'_k - E_k|) = \frac{1}{\sqrt{2\pi \delta}} e^{-\frac{(|E'_k - E_k| - \omega_R)^2}{2\delta^2}}, \tag{75}
\]

where the pre-factor \( e^{-i(\omega_1^L + \cdots + \omega_N^L) t} = 1 \) holds, and we can take \( t \to \infty \) without problem. If the resonance peak is very sharp, we are allowed to approximate the Gaussian distribution at its peak value,

\[
W(\omega_k^L) \approx \frac{1}{\sqrt{2\pi \delta}}, \text{ with the condition } E'_k = E_k \pm \omega_R. \tag{76}
\]

Now, we are ready to include the factors \( K(z, z') \)'s, being ignored in the above, and to obtain the final formula of transition amplitude in QO.

The formula for the transition amplitude \( \langle S(t, -\infty) \rangle_{\omega_L} \) for \( t = \infty \), or the S-matrix \( \langle S(\infty, -\infty) \rangle_{\omega_L} \) in QO is obtained as follows:

\[
\langle E_f | \hat{S} | E_i \rangle = 1 + \sum_{N=1}^{\infty} \left( -\frac{2\pi i}{\hbar \sqrt{2\pi \delta}} \right)^N \prod_{k=1}^{N} \int_{-L/2}^{L/2} dz_1 dz_2 \cdots dz_k \ K(z, z_{(P_D,C_1}) (E_f | \hat{H}_1(0) | E_i) \times K_{C_1;C_2}(z_1, z_2) K_k^L(z_1, z_2) \langle E'_2 | \hat{H}_1(0) | E_2 \rangle K_{C_2;C_3}(z_2, z_3) K_k^L(z_2, z_3) \times \cdots \times K_{C_N;C_0}(z_{N-1}, z_N) K_k^L(z_{N-1}, z_N) \langle E'_N | \hat{H}_1(0) | E_1 \rangle K_{C_N;C_0}(z_N, z_0), \tag{77}
\]

where the energy difference can be taken as \( E'_k - E_k = \pm \omega_R \) at each vertex, and \( \delta/2 \) is the width of laser frequency distribution around the resonance point \( \omega_R \). The factors \( K \)'s are given in Eqs.\ref{eq:75}--\ref{eq:79}, and the vertices \( \langle E'_f | \hat{H}_1(0) | E \rangle \) are in Eq.\ref{eq:76}, being categorized into six types, \((4)\pm_4, (4)\pm_2 \) and \((2)\pm_2 \).

Before ending this section, let us summarize the expressions for \( K \)'s and vertices:

The damping factors \( K \)'s are

\[
K = \begin{cases} 
\bar{K}_{C;C'}^L(z, z') = \frac{e^{-\gamma(z - z')}}{1 - r_{e^{i\phi}(L\ell)}}; \\
K_{C}^L(z, z') = \frac{1}{1 - r_{e^{i\phi}(L\ell)}}; \\
K_{P_D;C_1}^L(z, z_f) = \sqrt{1 - r_1^2 e^{-i\phi(L\ell)}} \frac{e^{-\gamma(t/2 - z_f)}}{1 - r_{e^{i\phi}(L\ell)}};
\end{cases} \tag{78}
\]

where \( r_{\omega}, \omega_L = (r_1 r_2 r_3 r_4) \omega, \omega_L \), and \( \phi(\omega L / c) = \frac{\omega}{c}(L - \ell) + \frac{\omega}{c} \ell \) (\( \ell \): the length of the non-linear crystal, \( L \): the circumference of the ring resonator).

\[\text{Here, we have note that in doing the contour integral in the complex plane of } \omega_k^L, \text{ no other poles appear, even if the factor } K \text{'s are included, since the denominator of the factor } K_{C;C'}^L(z, z') \text{ is } 1 - r_{e^{i\phi}(L\ell)}, \text{ and it can not be zero, unless the mirror perfectly reflects the laser beam } (r_L < 1).\]
The vertices $V$’s are

\begin{align}
(4)_4: \  & \langle n_1, n_2 | \hat{H}_1(0) | n_1 - 4, n_2 \rangle = 10 \varepsilon_0 \chi^{(4)} \left( \frac{\hbar \omega L}{4L^2} \right)^2 \alpha_{\text{cryst}} E_L \sqrt{(n_1 - 3)(n_1 - 2)(n_1 - 1)n_1}, \\
(4)_{-4}: \  & \langle n_1, n_2 | \hat{H}_1(0) | n_1 + 4, n_2 \rangle = 10 \varepsilon_0 \chi^{(4)} \left( \frac{\hbar \omega L}{4L^2} \right)^2 \alpha_{\text{cryst}} E_L \sqrt{(n_1 + 1)(n_1 + 2)(n_1 + 3)(n_1 + 4)}, \\
(4)_2: \  & \langle n_1, n_2 | \hat{H}_1(0) | n_1, n_2 - 2 \rangle = -10 \varepsilon_0 \chi^{(4)} \left( \frac{\hbar \omega L}{4L^2} \right)^2 \alpha_{\text{cryst}} E_L (2n_1 + 1) \sqrt{n_2(n_2 - 1)}, \\
(4)_{-2}: \  & \langle n_1, n_2 | \hat{H}_1(0) | n_1, n_2 + 2 \rangle = -10 \varepsilon_0 \chi^{(4)} \left( \frac{\hbar \omega L}{4L^2} \right)^2 \alpha_{\text{cryst}} E_L \sqrt{(n_2 + 2)(n_2 + 1)}, \\
(2)_2: \  & \langle n_1, n_2 | \hat{H}_1(0) | n_1, n_2 - 2 \rangle = 3 \varepsilon_0 \chi^{(2)} \left( \frac{\hbar \omega L}{4L^2} \right) \alpha_{\text{cryst}} E_L \sqrt{(n_2 - 1)n_2}, \\
(2)_{-2}: \  & \langle n_1, n_2 | \hat{H}_1(0) | n_1, n_2 + 2 \rangle = 3 \varepsilon_0 \chi^{(2)} \left( \frac{\hbar \omega L}{4L^2} \right) \alpha_{\text{cryst}} E_L \sqrt{(n_2 + 2)(n_2 + 1)}.
\end{align}

(79)

Now, the general formula has been obtained, which is useful in quantum optics (QO) to estimate the higher-order effects inside the ring resonator with a non-linear material contained.

### 6 Conclusion and Discussion

We intend in this paper to derive a general formula to be used to estimate the higher-order effects in QO, especially in a ring resonator with a non-linear material contained. The obtained formula is expressed Feynman graphically using propagators and vertices, in which the propagators include the effects of damping in the non-linear material as well as the reflection and transmission by mirrors.

We will discuss the possible observables being calculable in principle in the above-mentioned QO.

1. **Average number of produced photons:**
   We consider a simple case of NL-material with non-linear susceptibility, $\chi^{(2)}$ and $\chi^{(4)}$. For the laser beam with a resonance energy $\omega^L$, two photons $\gamma_1$ and $\gamma_2$ are relevant, having energies $\omega^L/4$ and $\omega^L/2$, respectively. We can estimate the averaged number of photons $\langle n_1 \rangle$ and $\langle n_2 \rangle$ for $\gamma_1$ and $\gamma_2$, as well as the correlations, $\langle (n_1)^m(n_2)^n \rangle$, $(m, n) = 1, 2, \cdots$. To do this we have to use the numerical simulations, and to compare the result so obtained with a theoretical study of the “generalized squeezing” in $\text{[1]}$.

2. **Husimi function:**
   Averaged number of the produced photons is directly connected to the Husimi function $H(\alpha)$ $\text{[9]}$. (Please refer to Wigner function $\text{[10]}$.) It is a quantum version of the distribution function and is called “Q function” $Q(\alpha)$ in QO:

$$H(t, \alpha) \equiv Q(t, \alpha) = \langle \alpha | \hat{\rho}(t) | \alpha \rangle,$$

(80)

where $\hat{\rho}(t)$ is a density matrix at time $t$, and $| \alpha \rangle$ is a coherent state with a complex eigen-value $\alpha$ for the annihilation operator $\hat{a}$.

In our case, we have two sets of annihilation operators, so that the Husimi function can be written at $t = \infty$ in terms of $S$-matrices as follows:

$$H(\alpha_1, \alpha_2) = \langle \alpha_1, \alpha_2 | \hat{S}|0\rangle \langle \alpha_1, \alpha_2 | \hat{S}|0\rangle^* = e^{-(|\alpha_1|^2 + |\alpha_2|^2)}$$

$$\times \sum_{\{\tilde{n}_1, \tilde{m}_1, \tilde{n}_2, \tilde{m}_2\} = 1}^{\infty} \frac{(\alpha_1^{2\tilde{n}_1}(\alpha_1)^{2\tilde{m}_1}(\alpha_2^{2\tilde{n}_2}(\alpha_2)^{2\tilde{m}_2})}{(2\tilde{n}_1)!(2\tilde{m}_1)!(2\tilde{n}_2)!(2\tilde{m}_2)!} \times \langle 2\tilde{n}_1, 2\tilde{n}_2 | \hat{S}|0\rangle \langle 2\tilde{m}_1, 2\tilde{m}_2 | \hat{S}|0\rangle^*,$$

(81)

(3) Observable to measure beyond the squeezing:
If we introduce a phase shifted wave,
\[ E^\theta(t, z) = e^{-i\theta} E^{-i\omega_L(t-z/c(z))} + e^{i\theta} E^{i\omega_L(t-z/c(z))}, \] (82)
then, it can accommodate the vector potential \( A(t, z) \propto E^{\theta=\pi/2}(t, z) \) and the electric field \( E(t, z) = E^{\theta=0}(t, z) \), which are canonical conjugate with each other. Therefore, the correlation functions of the phase sifted waves described the squeezing.

In our case, there exist various two-point and three-point correlation functions, since the decay of \( \gamma_2 \to \gamma_1 + \gamma_1 \) is possible by \( \omega_2 = 2\omega_1 \),

\[
\begin{cases}
\langle \Delta E_1^\theta(t) \Delta E_1^\theta(t') \rangle_{PD} = C_{11}^\theta(t, t'), \\
\langle \Delta E_2^\theta(t) \Delta E_2^\theta(t') \rangle_{PD} = C_{22}^\theta(t, t'), \\
\langle T[\Delta E_2^\theta(t_1) E_1^\theta(t'_1) \Delta E_2^\theta(t_2)] \rangle_{PD} = C_{12}^\theta(t_1, t_1', t_2),
\end{cases}
\] (83)

the last three-point correlation function gives, by specifying the ordering of time, as

\[
\begin{cases}
\langle \Delta E_2^\theta(t_1) E_1^\theta(t'_1) \Delta E_2^\theta(t_2) \rangle_{PD} = C_{112}^\theta(t_1, t'_1, t_2) \text{ for } (t_1 > t'_1 > t_2), \\
\langle \Delta E_1^\theta(t_1) \Delta E_2^\theta(t_2) E_1^\theta(t'_1) \rangle_{PD} = C_{121}^\theta(t_1, t_2, t'_1) \text{ for } (t_1 > t_2 > t'_1), \\
\langle \Delta E_2^\theta(t_2) \Delta E_1^\theta(t_1) \Delta E_2^\theta(t'_1) \rangle_{PD} = C_{211}^\theta(t_2, t_1, t'_1) \text{ for } (t_2 > t_1 > t'_1).
\end{cases}
\] (84)

Corresponding to the above correlation functions, we can introduce various spectra:

\[
\begin{align*}
S_{11}(\nu, \theta) &= \int_0^\infty d\tau C_{11}^\theta(t, t' = t + \tau)e^{-i\nu\tau}, \\
S_{22}(\nu, \theta) &= \int_0^\infty d\tau C_{22}^\theta(t, t' = t + \tau)e^{-i\nu\tau}, \\
S_{112}(\mu, \nu, \theta) &= \int_0^\infty d\tau \int_0^\infty d\sigma C_{112}^\theta(t_1, t'_1 = t_1 + \tau, t_2 = t_1' + \sigma)e^{-i(\mu\tau + \nu\sigma)}, \\
S_{121}(\mu, \nu, \theta) &= \int_0^\infty d\tau \int_0^\infty d\sigma C_{121}^\theta(t_1, t_2 = t_1 + \tau, t'_1 = t_1' + \sigma)e^{-i(\mu\tau + \nu\sigma)}, \\
S_{211}(\mu, \nu, \theta) &= \int_0^\infty d\tau \int_0^\infty d\sigma C_{211}^\theta(t_2, t_1 = t_2 + \tau, t'_1 = t_1' + \sigma)e^{-i(\mu\tau + \nu\sigma)}.
\end{align*}
\] (85)

The second spectrum \( S_{22}(\nu, \theta) \) was measured by L-A Wu et al.\[3\] using the OPO which contains the non-linear crystal with non-vanishing \( \chi^{(2)} \). This measurement shows the squeezing is realized in OPO. The spectrum \( S_{22}(\nu, \theta) \) represents the down-conversion of the laser photon to two degenerate photons \( (\omega_L \to 2\omega_2) \).

Therefore, it is interesting to examine the new spectra \( S_{112}(\mu, \nu, \theta) \), \( S_{121}(\mu, \nu, \theta) \) and \( S_{211}(\mu, \nu, \theta) \), in addition to \( S_{11}(\nu, \theta), S_{22}(\nu, \theta) \), in case of NL-crystal with non-vanishing \( \chi^{(2)} \) and \( \chi^{(4)} \).

We hope to study these issues in the next paper of our [11].

References

[1] “Introduction to Quantum Optics” by G. Grynberg, A. Aspect and C. Farbre, Cambridge University Press, Cambridge (2010).
“Quantum Optics” (in Japanese) by E. Hanamura, Iwanami Pub. Co., Tokyo (1991).

[2] S. Iso, “Quantum Field Theory as the Basis of Modern Physics” (in Japanese), Kyoritsu Pub. Co., Tokyo (2015).

[3] L-A Wu, H. J. Kimble, J. L. Hall, and H. Wu, Phys. Rev. Lett. 57, 2520 (1986); L-A Wu, Min Xiao, H. J. Kimble, Phys. Rev. Lett. 57, 2520 (1986), J. Opt. Soc. Am. B 4, 1465 (1987).
[4] S. Katagiri, A. Sugamoto, K. Yamaguchi and T. Yumibayashi, PTEP 2019, 12, 123B04 (2019).

[5] S. L. Braunstein and R. I. McLachlan, Phys. Rev. A 35, 1659 (1987), (Padé approximation);
O. B. Zaslavskii, Yu. A. Sinitsyn, and V. M. Tsukernik, Zh. Eksp. Teor. Fiz. 91, 156 (1986), Sov. Phys. JETP 64, 90 (1986).

[6] T. Kinoshita, J. Math. Phys. 3 650 (1962);
T. D. Lee and M. Nauenberg, Phys. Rev. D133 1549 (1964).

[7] Xing Fan, Shusei Kamioka, Kimiko Yamashita, Shoji Asai, Akio Sugamoto, PTEP 2018 (2018) no.6, 063B06.

[8] T. D. Lee and M. Nauenberg, Phys. Rev. D 133 1549 (1964).

[9] Kôdi Husimi, Proc. Phys. Math. Soc. Japan 22 264 (1940).

[10] E. P. Wigner, Phys. Rev. 40 749 (1932).

[11] [OUJ Tokyo Bynkyo Field Theory Collaboration] N. Aibara, N. Fujimoto, S. Katagiri, A. Sugamoto, K. Yamaguchi and T. Yumibayashi, in preparation.

[12] R. A. Fisher, M. M. Nieto, and V. D. Sandberg. “Impossibility of naively generalizing squeezed coherent states”, Phys. Rev. D 29.6 (1984): 1107.

[13] P. V. Elyutin, and D. N. Klyshko. “Three-photon squeezing: exploding solutions and possible experiments,” Phys. Lett. A 149.5-6 (1990): 241.

[14] R. A. Brandt and O. W. Greenberg. “Generalized Bose operators in the Fock space of a single Bose operator,” J. of Math. Phys. 10.7 (1969): 1168.

[15] G. d’Ariano, M. Rasetti, and M. Vadacchino, “New type of two-photon squeezed coherent states,” Phys. Rev. D 32.4 (1985): 1034.

[16] V. V. Dodonov, “Nonclassical states in quantum optics: a ‘squeezed’ review of the first 75 years,” J. of Optics B: Quantum and Semiclassical Optics 4.1 (2002): R1.

[17] S. L. Braunstein and C. M. Caves. “Phase and homodyne statistics of generalized squeezed states,” Phys. Rev. A 42.7 (1990): 4115.

[18] K. Banaszek and P. L. Knight. “Quantum interference in three-photon down-conversion,” Phys. Rev. A 55.3 (1997): 2358.

[19] L. C. G. Govia, E. J. Pritchett, and F. K. Wilhelm. “Generating nonclassical states from classical radiation by subtraction measurements,” New J. of Phys. 16.4 (2014): 045011.

[20] M. Cooper et al. “Experimental generation of multi-photon Fock states,” Optics express 21.5 (2013): 5309-5317.
Appendix

On the normalizablity of the generalized squeezing state, Fisher, Nieto and Sandberg \[12\] shows that the vacuum expectation value of the generalized squeezed state diverges and the perturbative calculations are broken. Elyutin and Klyshko \[13\] indicates that the state diverges in a finite time and behaves very differently from the normal squeezing state. To get around this problem, k-photon generalized boson operator was studied \[14, 15\] which uses the following operators \[16\]:

\[
\hat{A}(k) = \left( \left( \frac{n-k}{\hat{n}!} \right) \right)^{1/2} (\hat{a}^\dagger)^k, \quad \text{and} \quad \hat{S}(k) = \exp \left( z\hat{A}(k)^\dagger - z^*\hat{A}(k) \right),
\]

(86)

where

\[
\hat{n} = \hat{a}^\dagger\hat{a}, \quad [\hat{n}, \hat{A}(k), \hat{A}(k)^\dagger] = 1, \quad [\hat{n}, \hat{A}(k)] = -k\hat{A}(k),
\]

(87)

and \(\left[ \left[ \frac{n}{k} \right] \right]\) denotes the maximal integer less than or equal to \(\frac{n}{k}\).

On the other hand, Braunstein and McLachlan \[5\] argues that the divergence of the vacuum expectation value is a mere mathematical artifact, and compute it numerically using the Padé approximation. Later, Braunstein and Caves \[17\] calculates statistics on \(\hat{A}_k\) by homodyne and heterodyne measurements. Banaszek and Knight \[18\] computes the Wigner function about \(k = 3\) and shows that there is a negative region in its value. Govia, Pritchett and Wilhelm \[19\] proposes a method to generate generalized squeezed states from coherent states using Josephson photomultiplier (JPM).

Experimentally, Cooper et al. \[20\] generated a multiphoton Fock state. Recently, Chang et al. \[21\] observed the distribution of star states, which have been expected to be observed for \(k = 3\).