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# Cooperative properties of multiple quantum scattering: I quantum nutation

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E-mail: [enakinicolae@yahoo.com](mailto:enakinicolae@yahoo.com)**Keywords:** multiple cooperative scattering, quantum nutation in multi-step scattering, losses from cavities in multiple scattering

## Abstract

The cooperative models of the bimodal field in the multiple quantum scattering nutations are discussed and proposed for possible detections in open cavities. We proposed two types of cooperation between the converted photon processes in these multiple steps of scattering nutation in the cavity. One of them takes into consideration the cooperative process between the photons of each step of the multiple steps of Raman conversion. The second cooperative process takes place between the photons belonging to different steps of multiple scattering conversions. The proposed novel bimodal entangled sources take into consideration both the coherence and collective phenomena between the photons belonging to the system of the bimodal field obtained in multiple scattering emissions. The application of higher-order multiple Raman bimodal coherent field in quantum information is proposed.

## 1. Introduction

Cooperative processes of the simultaneous multi-wavelength emissions open new perspectives in the development of modern communication systems like holography and phase correlations, in the description of the interaction of light with biomolecules and living systems, taking into consideration not only classical aspect of these problems [1–10], but their quantum interpretation too [11–13]. The classical aspects of multi-photon stimulation coherent emission already are in the potential applications in registration medias, medical instrumentation, laser spectroscopy, LIDAR, and nonlinear optical mixers [1, 14–19]. Recently, specific attention is given to the new type of coherent emissions, which occur not only among the same quanta but between the photon groups generated in the nonlinear interaction of the electromagnetic field (EMF) with emitters (atoms, molecules, biomolecules, etc.). The quantum aspects of this type of emission were intensively studied in [11–13, 20–22], but multiple conversions of the photons and their quantum correlations remain today in the development studies. This type of light generation supports the idea of coherent correlation that appears in the bi-modal field, in which the entangled photons are generated. A physical characteristic of the radiation formed from the blocks of well-correlated bi-modes must be determined by the intensity of the electric field of each mode and propriety in such superposition. An attractive aspect of the problem consists in the selective two-quantum excitation of some atoms, or molecules of the system, where it is necessary to minimize the dipole active radiation in comparison with Raman emission. The last idea can be applied in microbiology [13, 23], where a selective dis-activation of some molecular structures (for example of viruses) in the tissue may become possible in induced Raman excitation. In such situations appears a necessity for a good description of both the amplitude and phase of this new type of radiation formed from bimodal correlated photons.

The intensity correlatives between the adjacent modes of Raman scattering and two-photon lasing were studied, [24]. Here we discussed the mutual interaction between two lasing processes. First process is described by single step induced scattering and second one takes into consideration two-photon induce processes, in which we have taken into consideration the quantum correlations between them. As multi-steps aspects of the scattering were not studied in the literature, we propose below to discuss the possible quantum correlations between the scattered modes in multistep process of quantum nutation. It is proposed two types of cooperative effects between the converted photons in these multiple steps induce process. The first type involves the

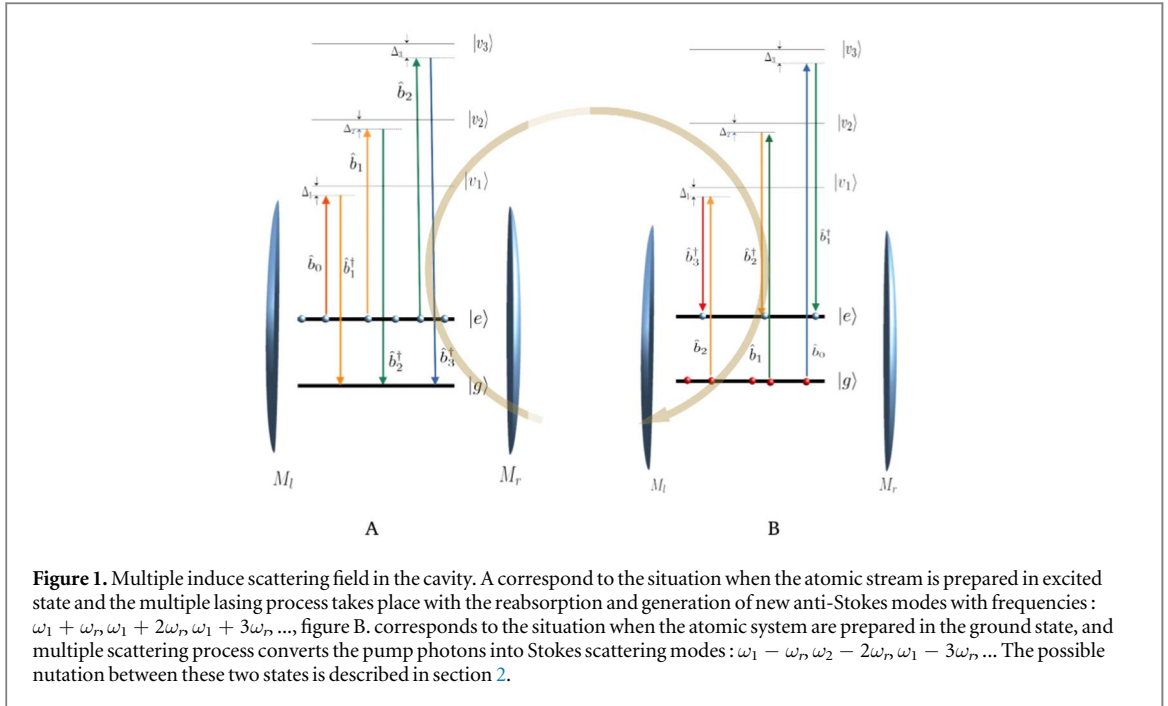
cooperative process between the photons of one of the Raman conversion steps. The second cooperative effect appears between the photons belonging to different steps of multiple conversions. This new type of generated coherent state is proposed by novel bimodal entangled sources, which take into consideration both the coherence and collective phenomena between the photons belonging to the system of the bimodal field obtained in multiple Raman emissions. The application of higher-order multiple Raman bimodal coherent field in quantum information is described in accordance with the definition of quantum amplitude and phase of such entangled states of light.

The correlation between the proposed model and experiment may be found in the modern experiments. For example the induced correlations stimulated by scattering conversion with the single and multiple Raman shifts were observed in KTP crystal pumped by a *Nd: YVO<sub>4</sub>* 1064-nm laser in [1]. The first-Stokes emission at wavelength 1095 nm was observed at Raman shift of 266 cm<sup>-1</sup> and emission power about 0.81 W stimulated by pump power of 16.5 W. The second output coupled with high reflectivity in the range of 1000 – 1130 nm is employed to simultaneously generate the first-Stokes 1095 nm and the second Stokes 1128 nm emissions of the same Raman shift about 266 cm<sup>-1</sup>. Similar phase-matched signals are observed in a *TiO<sub>2</sub>* crystal, in which the high-order coherent anti-Stokes Raman scattering signals of the fundamental light ( $\omega_p, k_p$ ) at frequencies and wave vectors,  $\omega_p + l\omega_R$  and  $k_p + lk$  respectively, where multiple Raman scattering number,  $l$ , reaches 10 under the optimum condition of phase-matching [25, 26]. Here  $\omega_R \sim |q| = 610\text{cm}^{-1}$  is the wave number of a Raman-active, A<sub>1g</sub>, mode of *TiO<sub>2</sub>*. This procedure of generation of the high-order harmonic conversion of infrared and visible light into extreme ultraviolet or soft x-ray, and high-order stimulated Raman scattering was in the center of attention of many experimental and theoretical investigations [2–10]. For example, in the [4–6, 27] a Raman spectrum with a large bandwidth was observed, ranging from the IR to the UV. Under some conditions, strong pump depletion was observed and up to five anti-Stokes sidebands were observed to have energies exceeding 10% of the transmitted pump pulse energies.

The quantum description of the multiple scattering process in which the atomic ensemble enters the quantum nutation process relative to multiple photon conversions is given in section 2. The possibilities to solve this problem for a relatively small number of radiators and a big number of scattered photons are proposed. The inverse solution, which is considered a small photon number and a big number of atoms, is given introducing the symmetries between the multiple steps of conversion. A new cooperative number that takes into consideration the number of photons and number of conversion steps is introduced. Considering that in the dielectric cavities the radiators are placed in the evanescent zone of the sphere/fiber in sections 3 and 3 we open the system of atoms in multiple scattering, introducing the interaction of these radiators with external EMF. The rate equations for the numbers of converted photons and excitation ones were obtained. In order to understand the possibilities of experimental observation of quantum correlations among different mode components of the cavity in the multiple scattering processes, the description of the higher order rates of the photons from such a system and their fluctuations are introduced. In section 4 we propose the algorithm of system of wave vectors in which multiple scattering processes took place and in appendix this system of vectors was constructed. It gives us the possibility of the description of the correlation of photons from non-adjacent steps of multiple scattering process. On the bases of this vector system, a master equation for the quantum generator with multiple scattering processes take place is proposed in section 3. Cooperative correlations between the photons which belong to the bimodal field of the multiple scattered processes were analytically estimated and numerically represented. New cooperative aspects between cavity photons belonging to the bimodal field were established and annualized in discussions 4 and conclusions 5.

## 2. Symmetry of multiple scattering transitions and their cooperative description

We represent higher-order multiple Raman conversion of photons by excited two-level radiators (atoms, excitation, molecules, molecular vibrations, etc). Taking into consideration the position of virtual states  $|v_1\rangle, |v_2\rangle$  and  $|v_n\rangle$ , we propose to study the opportunity of quantum nutation process between two-level atomic system and cavity excitation during the conversion of pump photons into multiple bimodal cavity modes of photons from the pump field into  $n$  scattered modes (see figure 1). In figure 1 we represent a possible three steps of multiple scattering of photons into new anti-Stokes modes. We will demonstrate the possibility to achieve the total conversion of all photons in the higher energy mode, represented in figure 1(B), and the possible return of the system back to its initial state during the quantum nutation which includes three types of the cooperative phenomena. The first type corresponds to Dicke cooperative process of  $N_r$  radiators, the second contains the cooperative process between the photons converted between the two modes in the same scattering step [22, 24], and the third cooperative process includes the effects of re-absorption of scattered photons and their conversion in the next steps of multiple scattering described in section 1.



According to perturbation theory, the transition matrix element in each step is proportional to the product of dipole active transition elements from ground and excited states to the nearest virtual one and inverse proportional to the detuning from resonances, with virtual state,  $\Delta_p$ ,  $g_p \sim d_{ev_p} d_{v_p g} q_{p-1} q_p / (\hbar^2 \Delta_p)$ . Here  $d_{ei}$  and  $d_{ig}$  are the dipole transition matrix elements from excited/ground to intermediary state,  $|v_p\rangle$ ;  $q_{p-1}$  and  $q_p$  are the per photon strength of the two adjacent cavity modes,  $p-1$  and  $p$  of the cavity. In order to simplify the multiple scattering problem, we will reduce the symmetry of multiple scattering process to two well known in quantum optics  $su(2)$  and  $su(1, 1)$  algebras. The Hamiltonian of the such two-level system connects bimodal cavity field in the higher order Raman scattering process and can be represented through the free,  $\hat{H}_{C0}$ , and interaction,  $\hat{H}_I$ , parts by the expressions,

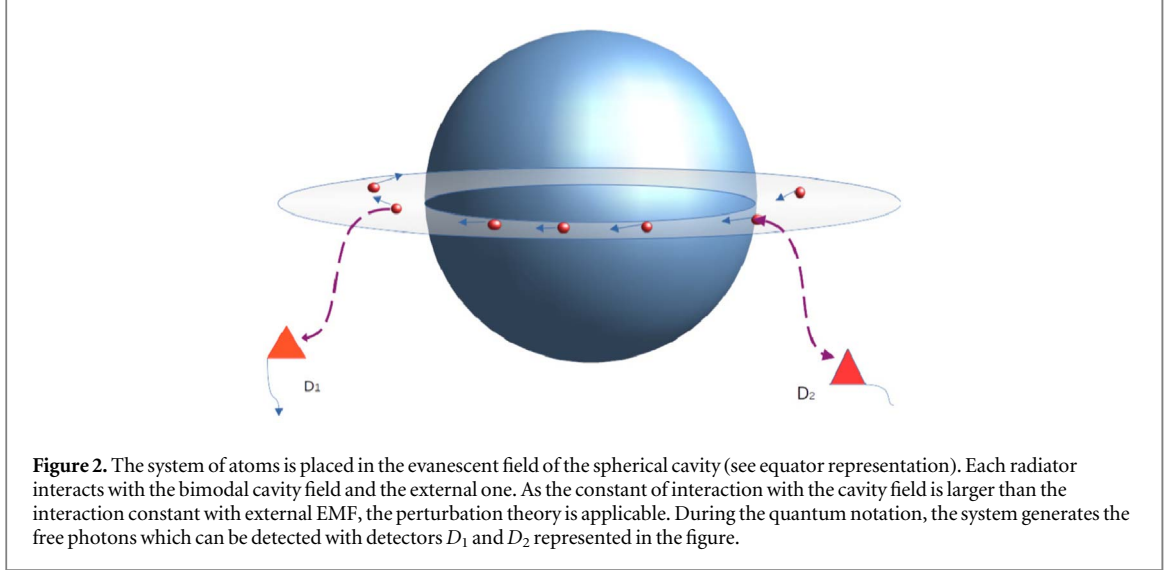
$$\hat{H}_C = \hat{H}_{C0} + \hat{H}_I, \quad (1)$$

$$\hat{H}_{C0} = \hbar \sum_{p=0}^n \omega_p \hat{c}_p^\dagger \hat{c}_p + \hbar \omega_r \hat{D}_z, \quad (2)$$

$$\hat{H}_I = \hbar \chi_n \{ \hat{\Lambda}_n^- \hat{D}^+ + \hat{\Lambda}_n^+ \hat{D}^- \}. \quad (3)$$

Here  $\hat{\Lambda}_n^- = \sum_{p=0}^{n-1} g_{p+1} \hat{c}_{p+1}^\dagger / \chi_n$ , and  $\hat{\Lambda}_n^+ = \sum_{p=0}^{n-1} g_{p+1} \hat{c}_p \hat{c}_{p+1}^\dagger / \chi_n$  are the annihilation and creation operators in the higher order of Raman scattering. Such multistep scattering processes take place from excited states of two level system in which the pump field is described by annihilation,  $\hat{c}_0$ , and creation,  $\hat{c}_0^\dagger$  operators. The scattering process in the  $p$ -order anti-Stokes modes generated in the  $n$ -multiple Raman scattering is described by the same annihilation and creation operators,  $\hat{c}_p$ , and,  $\hat{c}_p^\dagger$ ,  $p = 1, 2, \dots, n$ . As a result the pump and scattered field operators satisfy the same Bose commutation rules:  $[\hat{c}_{p'}, \hat{c}_p^\dagger] = \delta_{j,i}$ , and  $[\hat{c}_{p'}, \hat{c}_p] = 0$ ,  $p', p \equiv 0, 1, 2, \dots, n$ . The lowering,  $\hat{D}^- = \sum_{j=1}^N \hat{D}_j^-$ , and excitation,  $\hat{D}^+ = \sum_{j=1}^N \hat{D}_j^+$ , atomic operators are connected with the inversion through the commutation relation,  $[\hat{D}^+, \hat{D}^-] = 2\hat{D}_z$ , and  $[\hat{D}_z, \hat{D}^\pm] = \pm \hat{D}^\pm$ , where  $\hat{D}_z = \sum_{j=1}^N \hat{D}_{zj}$ , is the inversion operators of this radiator subsystem. Here the atomic operators are superposition of each atomic operators,  $\hat{D}_j^-$ ,  $\hat{D}_j^+$ , and  $\hat{D}_{zj}$  from the ensemble of the  $N_r$  undistinguished atoms. The atomic collective polarization operator depends on the number of the number radiators,  $N_r$ , and can be expressed through the creation,  $\hat{d}^\dagger$ , and annihilation,  $\hat{d}$ , operators of the excited state of the ensemble of atoms according to Holstein-Primakoff representation:  $\hat{D}^+ = \hat{d}^\dagger \sqrt{N_r - \hat{d}^\dagger \hat{d}}$ , and  $\hat{D}^- = \sqrt{N_r - \hat{d}^\dagger \hat{d}} \hat{d}$ , and  $D_z = -N_r/2 + \hat{d}^\dagger \hat{d}$  (see for example the representations from [28–31]). We observe that for the big number of atoms in the ground state,  $N_r \gg \langle \hat{d}^\dagger \hat{d} \rangle$ , the atomic operators can be regarded as a boson creation and annihilation one  $\hat{D}^+ \simeq \hat{d}^\dagger \sqrt{N_r}$ ,  $\hat{D}^- \simeq \sqrt{N_r} \hat{d}$ .

We introduce the losses from the closed system ‘atoms + cavity field’ considering that the atomic subsystem is concentrated in the evanescent zone of a spherical cavity or fiber so that the radiators are in scattering interaction with the intrinsic cavity field (3) described by discrete modes,  $\omega_0, \omega_1, \omega_2, \dots, \omega_{N_r}$ , and scattering photons into external free EMF as this is represented in figure 2. The cavity photons prepared in the pump mode



are in the multiple Raman conversion process stimulated by the radiator subsystem prepared in an excited state. Taking into consideration the position of virtual states,  $|v_1\rangle$ ,  $|v_2\rangle$ , and  $|v_r\rangle$ , below we describe the possibility of conversion photon generated in the scattered cavity modes into external free one in the process of simultaneous interaction of the atom with the cavity and external quantified field. So we divide the Hamiltonian into two parts in which the first part corresponds to the closed subsystem described by Hamiltonian (1), and the second one is the Hamiltonian of external EMF which stimulates the losses from the cavity during the photon convention,

$$\hat{H}_T = \hat{H}_C + \hat{H}_B + \hat{H}_{BC}. \quad (4)$$

Here  $\hat{H}_C$  is the Hamiltonian of the radiator and cavity field subsystems defined by Exps. (2), and (3). The free part of Hamiltonian of external EMF,

$$\hat{H}_B = \hbar \sum_k \omega_k \hat{b}_k^\dagger \hat{b}_k,$$

can be combined with the interaction part when the atomic subsystem is prepared in the excited state,  $\hat{H}_{BC}^e = \sum_{k,p} \hbar \chi_{k,p} \{ \hat{c}_p \hat{b}_k^\dagger \hat{D}^- + H.c. \}$ . The excitation from ground states may be described by the term of the Hamiltonian term,  $\hat{H}_{BC}^g = \sum_{k,p} \hbar \chi_{p,k} \{ \hat{c}_p \hat{b}_k^\dagger \hat{D}^+ + H.c. \}$ , which is applicable for the atomic subsystem prepared in the ground state. Considering the two processes of scattering possible, when the atomic subsystem can pass into quantum notation we define the interaction with the external field as a sum of the both Hamiltonian parts,  $\hat{H}_{BC} = \hat{H}_{BC}^e + \hat{H}_{BC}^g$ .

The free field is described by the operators of emitted photons,  $\hat{b}_k^\dagger$  and  $\hat{b}_k$ , which leave the evanescent zone in the free space. These emitted photons don't take part in the next steps of the spontaneous scattering process. The number of such photons,  $\hat{N}_e = \sum_k \hat{b}_k^\dagger \hat{b}_k$ , and its statistics give us information about quantum notation between the cavity field and atomic subsystem trapped in the evanescent zone. For molecular vibration, described by the above boson operators, when the pump is very strong its field operator can be described by  $C$ - number operator the Schrödinger equation can be solved exactly [7, 8]. In this section, we use the quantum approach for the loss of photons from the system. Indeed, passing into rotation system of coordinate,  $|\psi(t)\rangle = T \exp[it\hat{H}_{T0}/\hbar] |\bar{\psi}(t)\rangle$ , we obtain the equation,

$$i\hbar \frac{\partial}{\partial t} |\psi(t)\rangle = \hat{H}_{IB}(t) |\psi(t)\rangle, \quad (5)$$

where,  $\check{H}_{IB}(t) = \exp[it\hat{H}_{T0}/\hbar] \hat{H}_{IB} \exp[-it\hat{H}_{T0}/\hbar]$ , and  $|\bar{\psi}(t)\rangle = \exp[-it\hat{H}_T/\hbar] |\psi(0)\rangle$ . Here  $\hat{H}_{IB} = H_I + H_{BC}$  is the interaction of radiators with cavity modes (3) and external field,  $\hat{H}_{T0} = \hat{H}_C + \hat{H}_B$  is the free Hamiltonian of subsystems in interaction picture,

$$\check{H}_{IB}(t) = \check{H}_I(t) + \sum_{k,p} \hbar \{ [\chi_{k,p} \check{c}_p^\dagger(t) \check{b}_k(t) + \chi_{p,k} \check{c}_p(t) \check{b}_k^\dagger(t)] \check{D}^+(t) + H.c. \}, \quad (6)$$

which describes the scattering process of cavity photons by atomic subsystems placed into the evanescent zone. The interaction constant,  $\chi_{k,p}$  is proportional to the same parameters of the system as  $g_p$ ,  $\chi_{k,p} \sim d_{evp} d_{vpg} q_k q_p / (\hbar^2 \Delta_p) q_k$  and  $q_p$  are the per photon strength of the cavity and free field modes. As these amplitudes are inversely proportional to the squirt from quantified volume,  $q_n \sim 1/\sqrt{v_0}$ , and  $q_k \sim 1/\sqrt{V}$ ,

where  $v_0$  and  $V$  are the volumes of the cavity and free space respectively, it follows that  $|\chi_{k,p}| \ll g_p$ . Considering that the Schrödinger equation (5) for closed cavity field  $\chi_{k,p}$  solvable, we introduce the operator,  $\hat{O}(t)$ , belonging to the cavity subsystem. For the mean value of this operator,  $\langle \hat{O}(t) \rangle = \text{Tr} \{ \hat{\rho}(0) \hat{O}(t) \}$ , we obtain the following Heisenberg equation,

$$\begin{aligned} \frac{d}{dt} \hat{O}(t) &= \frac{i}{\hbar} [\hat{H}_{0T}, \hat{O}(t)] \\ &+ i \sum_p \{ [\hat{c}_p^\dagger(t) \{ \chi_{k,p} \hat{D}^+(t) + \chi_{p,k} \hat{D}^-(t) \}, \hat{O}(t)] \hat{b}_k(t) \\ &+ H.c. \{ \hat{O}^\dagger(t) \rightarrow \hat{O}(t) \} \}. \end{aligned} \quad (7)$$

To eliminate the operators of the free field from this equation we use the traditional procedure of the representation of the solution of the annihilation,  $\hat{b}_k(t)$ , and creation Heisenberg operators,

$\hbar \chi_n \{ \hat{\Lambda}_n^- \hat{D}^+ + \hat{\Lambda}_n^+ \hat{D}^- \} \cdot \hat{b}_k^\dagger(t)$ , described in literature (see for example ([21, 22]). According to this procedure, the solution is represented through vacuum and sources parts,

$$\begin{aligned} \hat{b}_k(t) &= \hat{b}_k(0) \exp[-i\omega_k t] - i \sum_p \chi_{k,p} \int_0^t d\tau \exp[-i\omega_k \tau] \{ \hat{c}_p(t-\tau) \hat{D}^-(t-\tau) \\ &- i \sum_p \chi_{p,k} \int_0^t d\tau \exp[-i\omega_k \tau] \{ \hat{c}_p(t-\tau) \hat{D}^+(t-\tau) \}, \end{aligned} \quad (8)$$

so that vacuum part,  $\hat{b}_k^v(t) = \hat{b}_k(0) \exp[-i\omega_k t]$ , gives zero contribution in normal product of the right hand part of equation, (7),  $\hat{b}_k^v(t) |0\rangle_k = 0$ . The second term of solution (8) represent the source part in the Born-Markov approximation, the solution has the form,

$$\begin{aligned} \hat{b}_k(t) &= \hat{b}_k^v(t) - 2\pi i \sum_p \chi_{k,p} \hat{c}_p(t) \hat{D}^-(t) \xi(\omega_k - \omega_p - \omega_r) \\ &- 2\pi i \sum_p \chi_{p,k} \hat{c}_p(t) \hat{D}^+(t) \xi(\omega_k - \omega_p + \omega_r), \end{aligned} \quad (9)$$

where  $\xi(\omega) = \delta(\omega)/2 - iPv/(2\pi\omega)$  is the Fourier transform of the Heaviside step function. The solution for creation operator,  $\hat{b}_k^\dagger(t)$  is Hermit conjugate to the expressions (9). For the mean value of this operator,  $\langle \hat{O}(t) \rangle = \text{Tr} \{ \hat{\rho}(0) \hat{O}(t) \}$ , in the rotation wave approximation we obtain the following Heisenberg equation,

$$\begin{aligned} \frac{d}{dt} \langle \hat{O}(t) \rangle &- \frac{i}{\hbar} \langle [\hat{H}_C^r(t), \hat{O}(t)] \rangle \\ &= \sum_p \langle [\hat{c}_p^\dagger(t) \hat{D}^+(t), \check{O}(t)] \{ \Gamma_p^{(1)} \hat{c}_p(t) \hat{D}^-(t) + \Gamma_{p,p+2}^{(1)} \hat{c}_{p+2}(t) \hat{D}^+(t) \} \\ &+ \sum_p \langle [\hat{c}_p^\dagger(t) \hat{D}^-(t), \check{O}(t)] \{ \Gamma_p^{(2)} \hat{c}_p(t) \hat{D}^+(t) + \Gamma_{p,p-2}^{(2)} \hat{c}_{p-2}(t) \hat{D}^-(t) \} \\ &+ H.c. \{ \hat{O}^\dagger(t) \rightarrow \hat{O}(t) \}. \end{aligned} \quad (10)$$

Here the renormalized Hamiltonian,

$$\begin{aligned} \hat{H}_C^r &= \hbar \delta \hat{D}_z + \hbar \chi_n \{ \hat{\Lambda}_n^- \hat{D}^+ + \hat{\Lambda}_n^+ \hat{D}^- \} \\ &- \sum_p \{ \mathcal{E}_p^{(1)} \hat{D}^+ \hat{D}^- + \mathcal{E}_p^{(2)} \hat{D}^- \hat{D}^+ \} \hat{c}_p^\dagger \hat{c}_p \\ &- \sum_p \mathcal{E}_{p-1,p+1} \{ \hat{c}_{p+1}^\dagger \hat{c}_{p-1} \hat{D}^- \hat{D}^- + H.c. \}. \end{aligned}$$

is represented through the cooperative interaction shifts of energies between atomic and cavity modes stimulated by vacuum field,  $\mathcal{E}_p^{(1)} = \hbar \sum_k |\chi_{p,k}|^2 P v / (\omega_k - \omega_p - \omega_r)$ ,  $\mathcal{E}_p^{(2)} = \hbar \sum_k |\chi_{p,k}|^2 P v / (\omega_k - \omega_p + \omega_r)$  and energy correlation between the scattering steps  $\mathcal{E}_{p-1,p+2}^{(1)} = \hbar \sum_k \chi_{p-1,k} \chi_{k,p} + 1 P v / (\omega_k - \omega_{p+1} + \omega_r)$ . The cooperative losses of photons with frequencies for anti-Stokes,  $\omega_k = \omega_p + \omega_r$  and Stokes photons,  $\omega_k = \omega_p - \omega_r$ , are described by the expressions,  $\Gamma_p^{(1)} = \pi \sum_k |\chi_{p,k}|^2 \delta(\omega_k - \omega_p - \omega_r)$  and  $\Gamma_p^{(2)} = \pi \sum_k |\chi_{p,k}|^2 \delta(\omega_k - \omega_p + \omega_r)$ . The cooperative cross correlations between non-adjacent modes is described by losses,  $\Gamma_{p,p+2}^{(1)} = \pi \sum_k \chi_{p,k} \chi_{k,p+2} \delta(\omega_k - \omega_{p+2} + \omega_r)$ , and  $\Gamma_{p,p-2}^{(2)} = \pi \sum_k \chi_{p,k} \chi_{k,p-2} \delta(\omega_k - \omega_{p-2} - \omega_r)$ .

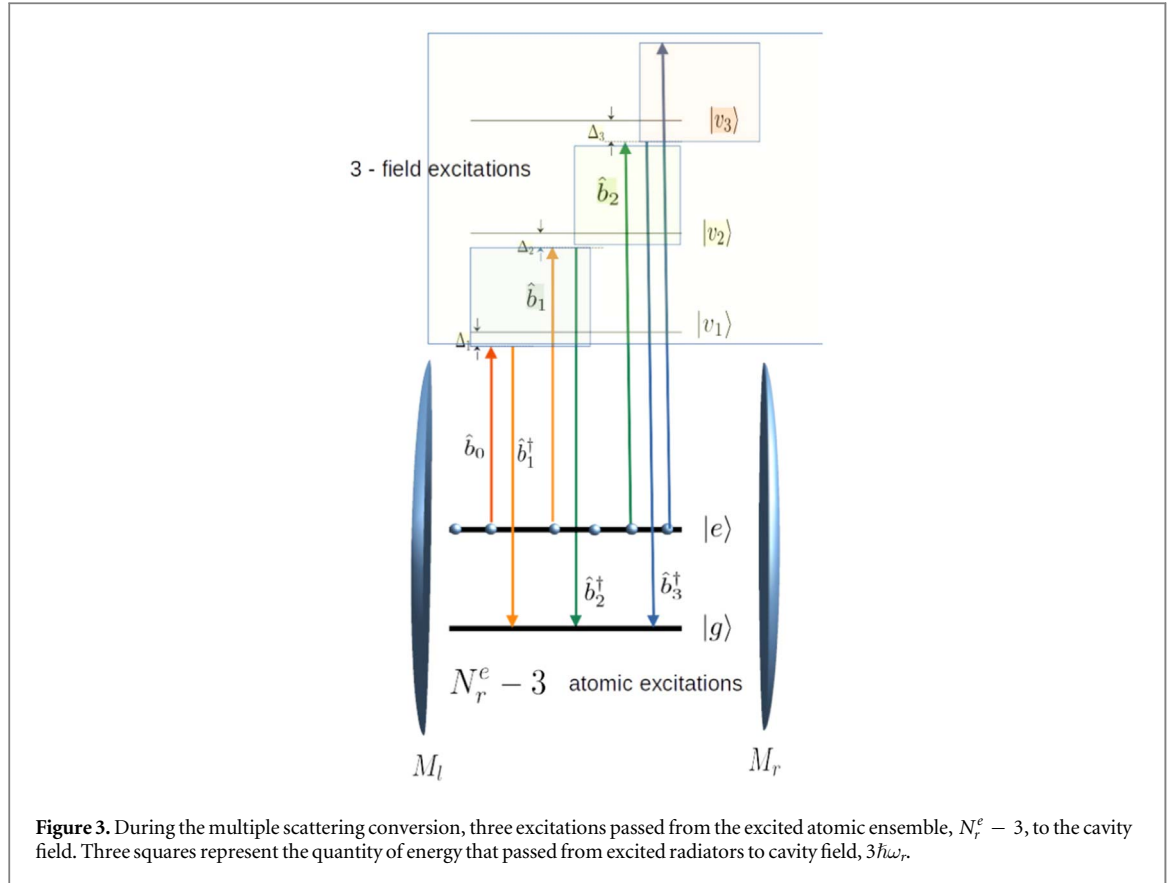
We observe from generalized equation (5) that the total number of photons in the system,

$\hat{N}_f(t) = \sum_{p=0}^n \hat{c}_p^\dagger(t) \hat{c}_p(t)$ , is not conserved and the emitted photon rate from the system ‘atoms+cavity field’

has the opposite sign relative the photon loses from the system,  $d\hat{N}_e(t)/dt = -d\hat{N}_f(t)/dt$ . From this follows that the number of emitted photons depends on the initial one in the pump field.  $N_f(0) = l$ ,

$\hat{N}_e = \sum_k \hat{b}_k^\dagger(t) \hat{b}_k(t) = N_f(0) - \hat{N}_f(t)$ , According to the generalized expression (10) the losses from system ‘C’ can be easy obtained by substitution of  $\hat{O}(t)$  with  $\hat{N}_f(t)$ . For the closed system, C, follows that it is also conserved





**Figure 3.** During the multiple scattering conversion, three excitations passed from the excited atomic ensemble,  $N_r^e - 3$ , to the cavity field. Three squares represent the quantity of energy that passed from excited radiators to cavity field,  $3\hbar\omega_r$ .

the total energy of excited atoms and field excitation in the cavity represented in figure 3 by squared figures. The total number of such excitation is,  $\hat{W}_C(t) = \sum_p p \hat{c}_p^\dagger(t) \hat{c}_p(t) + \hat{D}_z + N_r/2$ . We emphasize here that the expression,  $\hat{W}_f(t) = \sum_p p \hat{c}_p^\dagger(t) \hat{c}_p(t)$ , represents the number of cavity field excitations, which is obtained during the induced conversion of photons from one field mode to another. In this situation, we obtain the following expression for the loss of cavity excitations,

$$\begin{aligned} \frac{d}{dt} \langle \hat{W}_C(t) \rangle &= -2 \sum_{p=0}^{n-1} \Gamma_p^{(1)} (p+1) \langle \hat{c}_p^\dagger(t) \hat{c}_p(t) \hat{D}^+(t) \hat{D}^-(t) \rangle \\ &\quad - 2 \sum_{p=2} \Gamma_p^{(2)} (p-1) \langle \hat{c}_p^\dagger(t) \hat{c}_p(t) \hat{D}^-(t) \hat{D}^+(t) \rangle \\ &\quad - 2 \sum_{p=1} p \Gamma_{p-1,p+1} \{ \langle \hat{c}_{p-1}^\dagger(t) \hat{c}_{p+1}(t) \hat{D}^+(t) \hat{D}^+(t) \rangle + H.c. \}. \end{aligned} \quad (11)$$

The equation for the losses of the number of photons from the cavity,  $d\langle \hat{N}_f(t) \rangle / (dt)$  is obtained from Exp. (11) by substitution,  $\hat{W}_C(t) \rightarrow \hat{N}_f(t)$ , and  $(p+1) \rightarrow 1$  and  $(p-1) \rightarrow 1$  in the first and second term of expression (11). As follows from these definitions of the pump field with frequency,  $\omega_0$ , and  $p=0$ , we have the scattered field with frequencies:  $\omega_k = \omega_0 + \omega_r$ , for  $\Gamma_0^{(1)}$ ; and  $\omega_k = \omega_0 - \omega_r$ , for  $\Gamma_p^{(2)}$ . The last frequencies,  $\omega_k = \omega_0 - \omega_r$ , correspond to scattered pump field in the low frequency spectrum. If the system of atoms are prepared in excited state, and field in the lower frequency state,  $|l\rangle_0$ , the multiple scattering in frequencies,  $\omega_1, \omega_2, \dots, \omega_n$ , according to the interaction Hamiltonian (3) must give the maximal values of probabilities,  $|\chi_{p,k}|^2 \sim |g_p|^2$ , due to the small detuning from resonances,  $\Delta_p$ . Other frequencies,  $\omega_k < \omega_0$ , and  $\omega_k > \omega_n$ , are neglected from the model. In this situation the main contribution in Exp. (11) gives the first term proportional to  $\Gamma_p^{(1)} (p+1) \langle \hat{c}_p^\dagger(t) \hat{c}_p(t) \hat{D}^+(t) \hat{D}^-(t) \rangle$ . In opposite case when the atoms are prepared in the ground state (see figure 1), we can consider the pump field prepared in the state  $|l\rangle_n$  with maximal frequency,  $\omega_n$ , in which the pump field is accompanied by the downward scattering at the frequencies,  $\omega_{n-1} = \omega_n - \omega_r, \dots, \omega_0$ . The losses,  $\Gamma_n^{(1)}$ , at frequencies,  $\omega_k = \omega_n + \omega_r$ , must be eliminated from due to the fact that the upward scattering probability is considered smaller than downward one in the frequency interval  $\omega_0 < \omega_k < \omega_n$ . The quantum nutation possibilities between these two descriptions of scattering process is discussed below.

The observation of these quantities with the external detector may be realized introducing the number of emitted excitations in the external field,  $\hat{W}_e(t) = \sum_k [(\omega_k - \omega_0) / \omega_r] \hat{b}_k^\dagger(t) \hat{b}_k(t)$ . Here  $\hbar \sum_k (\omega_k - \omega_0) \hat{b}_k^\dagger(t) \hat{b}_k(t) = \hat{H}_B - \hbar \omega_0 \hat{N}_e$  is the excess of energy generated by open system,  $C$ , due to the multiple conversion process of the pump photons in

other modes (see figure 3). In this situation, we obtain the following expressions for the rate of emitted excitation  $d\hat{W}_e/dt$  and photon number

$$\frac{d}{dt}\langle\hat{W}_e(t)\rangle \simeq -\frac{d\langle\hat{W}_C(t)\rangle}{dt}; \quad \frac{d\langle\hat{N}_e(t)\rangle}{dt} = -\frac{d\langle\hat{N}_f(t)\rangle}{dt}. \quad (12)$$

Here for the conservation of the quantity,  $\hat{W}_C(t) + \hat{W}_e(t)$ , we neglect the dispersion of emitted photons in the expressions for the commutator  $[\hat{W}_C + \hat{W}_e, \hat{H}_{BC}] \simeq 0$ . We observe that this commutator is proportional to,  $\sum_{k,p} \hbar \chi_{k,p} [(\omega_k - \omega_0)/\omega_r - p - 1] \{\hat{c}_p^\dagger \hat{b}_k \hat{D}^+ - H.c.\}$ , from which follows that in the rotation wave approximation,  $\omega_k \simeq \omega_p + \omega_r = \omega_r(p + 1)$ , this commutator can be neglected.

Taking into consideration the conservation law of the moments of the total number of photons,  $[\sum_k \hat{b}_k^\dagger(t) \hat{b}_k(t)]^\alpha = [N_{ph}(0) - \hat{N}_{ph}(t)]^\alpha$ , and total excitation number,  $(\hat{W}_C)^\alpha$ ,  $\alpha = 1, 2, \dots$ , in the closed system,  $C$ , below we estimate the quantum moments of the losses rates,  $(d\hat{W}_C/dt)^\alpha$ , and  $(d\hat{N}_e/dt)^\alpha$ , of cavity quasi—quanta and photons from this system, when we introduce the interaction with external field. According to this representation we propose to estimate the quantum fluctuation of photon rate introducing the expression for square rate of quanta from the system 'C'. Defining the following normal product,

$$\begin{aligned} \langle :(\partial_0 \hat{W}_e(t))^2: \rangle &\sim \langle \partial_0 \hat{b}_k^\dagger(t) \partial_0 \hat{b}_{k'}^\dagger(t) \hat{b}_{k'}(t) \hat{b}_k(t) \rangle \\ &+ \langle \hat{b}_{k'}^\dagger(t) \partial_0 \hat{b}_k^\dagger(t) \partial_0 \hat{b}_{k'}(t) \hat{b}_k(t) \rangle + H.c., \end{aligned} \quad (13)$$

we can eliminate the operator of external field. Considering that the number of atoms in the excited state is larger than the number of atoms in ground one we obtain follow second moment (13),

$$\begin{aligned} \left\langle : \left( \frac{d}{dt} \hat{W}_e(t) \right)^2 : \right\rangle &= 2 \sum_{p,p'} \sum_{p_1,p'_1} \Gamma_{p,p'}(p' + 1) \{ \Gamma_{p_1,p'_1}(p'_1 + 1) \\ &\times \langle \hat{c}_p^\dagger(t) \hat{c}_{p_1}^\dagger(t) \hat{c}_{p'}(t) \hat{c}_{p'_1}(t) \hat{D}^+(t) \hat{D}^+(t) \hat{D}^-(t) \hat{D}^-(t) \rangle \\ &+ \Gamma_{p_1,p'_1}^*(p'_1 + 1) \langle \hat{c}_{p_1}^\dagger(t) \hat{c}_p^\dagger(t) \hat{c}_{p'}(t) \hat{c}_{p'_1}(t) \hat{D}^+(t) \hat{D}^+(t) \hat{D}^-(t) \hat{D}^-(t) \rangle \}, \\ &+ H.c., \end{aligned} \quad (14)$$

in which  $\Gamma_{p,p'} = 2\pi \sum_k \chi_{p,k} \chi_{p',k} \xi(\omega_k - \omega_{p'} - \omega_r)$  and  $\Gamma_{p,p'}^* = [\Gamma_{p,p'}(\omega_0 + p'\omega_r)]^*$ . According to the Exp. (14) the square fluctuation of the emission photon rate and excitation from the system can be estimated,

$$\begin{aligned} \Delta_N^2 &= \left\langle : \left( \frac{d}{dt} \hat{N}_e(t) \right)^2 : \right\rangle - \left\langle \frac{d}{dt} \hat{N}_e(t) \right\rangle^2, \\ \Delta_W^2 &= \left\langle : \left( \frac{d}{dt} \hat{W}_e(t) \right)^2 : \right\rangle - \left\langle \frac{d}{dt} \hat{W}_e(t) \right\rangle^2. \end{aligned} \quad (15)$$

The expression for the square intensity fluctuations,  $\Delta_W^2$ , can be easily obtained from the equation for losses (14), by replacing  $:(d\hat{W}_e(t)/dt)^2:$  with normal product  $:(d\hat{W}_e/dt)^2:$ .

Following the perturbation theory we can introduce the exact solutions of the Schrödinger equation for cavity Hamiltonian (1) in the right-hand side of the expressions (10), (11) and (14), considering that the interaction with the external field is quite low in computation with quantum Rabi nutation frequencies which described the closed cavity,  $\Gamma_p^{(i)}(p + 1) \ll \chi_n$ . In this situation, we may return to the generalized equation for arbitrary Heisenberg operator  $\hat{A}$ ,  $\langle \hat{A}(t) \rangle = \prod_k \langle 0_k | \langle \varphi_{l,n}(0) | \langle \psi_j(0) | \hat{A}(t) | \psi_j(0) \rangle | \varphi_{l,n}(0) \rangle | 0_k \rangle$ , representing it right hand side through the initial density matrix of the system,  $\hat{\rho}(0) = |\psi_j(0)\rangle \langle \psi_j(0)| \otimes |\varphi_{l,n}(0)\rangle \langle \varphi_{l,n}(0)| \otimes \prod_k |0_k\rangle \langle 0_k|$ ,

$$\begin{aligned} \langle \hat{A}(t) \rangle &= \sum_{j,m} \sum_{\{n_p\}} \sum_{\{n_k\}} \prod_{p=0}^n \prod_k \langle n_k | \langle n_p | \langle j, m | \hat{\rho}(0) \hat{U}^+(t) \exp[it(\hat{H}_C + \hat{H}_B)/\hbar] \\ &\times \hat{A}(0) \exp[-it(\hat{H}_C + \hat{H}_B)/\hbar] \hat{U}(t) | j, m \rangle | n_p \rangle | n_k \rangle_k, \end{aligned} \quad (16)$$

Here, under the trace, we have the Hamiltonian parts,  $\hat{H}_C$  and  $\hat{H}_B$ , described by the expressions (4) and (6). The initial state of atomic ensemble is prepared in excited state,  $|\psi_j(0)\rangle = |j, j\rangle$ , and photon subsystem is prepared in the  $l$  photon Fock state of low frequency mode,  $|\varphi_{l,n}(0)\rangle = |l\rangle_0 |0\rangle_1 \dots |0\rangle_n$ . The similar initial situation may be declared when the system of atoms are prepared in the ground state,  $|\psi_j(0)\rangle = |j, -j\rangle$ , while the bimodal cavity field in the higher frequency Fock state,  $|\tilde{\varphi}_{l,n}(0)\rangle = |0\rangle_0 |0\rangle_1 \dots |l\rangle_n$ . The evolution operator,  $\hat{U}(t)$ , can be represented through the  $T$  product of the interaction Hamilton (6),  $\hat{U}(t) = T \exp[-i \int_0^t dt' \hat{H}_{IB}(t')/\hbar]$ .

Passing from Heisenberg to Schrödinger picture in the expressions (10) and (16) we must solve exactly the Schrödinger equation of the closed system,  $C$ , and after that return to the right-hand side expressions for losses (11) and its moments. The bimodal field is considered so that the frequency deference,  $\omega = \omega_{p+1} - \omega_p$ , is constant



for each bimodal pair and the detuning from resonance,  $\delta = \omega_r - \omega \ll \omega_r$ , is relative smaller than radiator frequency,  $\omega_r$ . In the same short time approximation, taking into consideration the intrinsic symmetry between the bi-boson operator, we propose to solve the Schrödinger equation in interaction picture. Indeed, passing into rotation system of coordinate with frequency  $\omega$ , the Hamiltonian part,  $H_{0\omega} = \sum_{p=0}^n \omega_p \hat{c}_p^\dagger \hat{c}_p + \omega \hat{D}_z$ , commute with Hamiltonian of the ‘atoms + cavity field’ and becomes the motion integral. For the resonance case, this motion integral corresponds to the conservation of the total excitation number with energies in the resonance case,  $\delta = 0$ . The wave function in the rotation system of coordinate,  $|\psi_{j,l}(t)\rangle = \exp[it \{\sum_{p=0}^n \omega_p \hat{c}_p^\dagger \hat{c}_p + \omega \hat{D}_z\}] |\bar{\psi}_{j,l}(t)\rangle$ , is reduced to the Schrödinger equation,

$$i\hbar \frac{\partial}{\partial t} |\bar{\psi}_{j,l}(t)\rangle = \hat{H}_{Ief} |\bar{\psi}_{j,l}(t)\rangle, \quad (17)$$

where the effective interaction Hamiltonian takes into consideration the detuning from resonance relative bimodal scattering process,  $\hat{H}_{Ief} = \hbar\delta\hat{D}_z + \hbar\chi_n \{\hat{\Lambda}_n^- \hat{D}^+ + \hat{\Lambda}_n^+ \hat{D}^-\}$ , if the system of coordinates is rotated with frequency,  $\omega$ . The total number of photons,  $\hat{N}_{ph} = \sum_{p=0}^n \hat{c}_p^\dagger \hat{c}_p$ , commutes with operators,  $\hat{\Lambda}_n^-$  and  $\hat{\Lambda}_n^+$ . In other words, during the multiple scattering, the particles are converted from one spectral diapason to others, conserving their number. This conservation law is not the same as in traditional single photon interaction of the atomic ensemble with cavity field. In one scattering act the photon is converted from  $\omega_p$  to  $\omega_{p+1}$  and the field from the cavity received the energy,  $\hbar\omega = \hbar(\omega_{p+1} - \omega_p)$ . In the in single photon interaction in the transition event of the atom excited to ground state, the cavity field receives one photon with energy,  $\hbar\omega$ . The analogies between the coherent processes single photon interaction and multiple scattering field can be easily observed if we accept the number of above energy difference as a quasi-particle in a closed cavity. This suggests that the role of the number of photons in the multiple scattering process plays the operator,  $\hat{\Lambda}_z = \sum_{p=0}^n (\omega_p - \omega_0) \hat{c}_p^\dagger \hat{c}_p / \omega$ , which together with a number of excited atoms,  $\hat{N}_{ex} = N_r/2 + \hat{D}_z$ , conserve the total number of excitations in the closed ‘cavity+atoms’ system,  $\hat{\Lambda}_z + \hat{N}_{ex} = const$ , as this took place in the single photon interaction in the cavity [32]. From these analogies, we may introduce the operators,  $\hat{\Lambda}_n^-$  and  $\hat{\Lambda}_n^+$ , as an annihilation and creation of cavity quasi-particle with fixing energy,  $\omega_r = \omega_{k+1} - \omega_k$ . Initially, we can consider that all photons are in the pump mode with frequency,  $\omega_{p_0} \sim \omega_0$ , and during the multiple scattering is generated to such portions of energies in the resonator as this is represented in figure 3.

We can represent the solution of this equation in the two approach. First way correspond to the situation, when the excited subsystem of atoms is regarding as a pump flux which enter into the micro resonator in which the excited energy is converted into anti-Stokes modes in multiple scattering process of the special prepared pump mode into the resonator. The restoration of initial state of quantum system was studied from the statistical point of view in [33–35].

1. Let us start with quantum nuataion of the small ensemble of two - level system under multiple scattering with bimodal cavity field. In multiple photon conversions, the restoration probability of the initial state with increasing the number of atoms in the system and number of catering steps in the short time interaction with bimodal cavity field substantially decreases, due to multiple reabsorption and emission of pump photons. In the situation when the number of cavity photons is small we can define the  $N + 1$  vector operators of the atomic ensemble in the multiple Raman conversion of prepared pump mode described by the Hamiltonian (3) in the representation of the solution (17)

$$|\psi_{l,n}(t)\rangle = \sum_{m=-j}^j \alpha_m \hat{X}_m(t) |\varphi_{l,n}(0)\rangle, \quad (18)$$

where  $\{\hat{X}_m(t) = \exp[-i\hat{H}_{Ief}t] |j, m\rangle_r, m = -j, -l + 1, \dots, l - 1, j\}$  are vector—operators, which depends on the bimodal operators of cavity field,  $|\varphi_{l,n}(0)\rangle$ , is the initial state of the field in multiple scattered modes of the pump field. If we start from excited state, the initial coefficients are,  $\alpha_m = \delta_{m,j}$ . In this situation is better to consider that the initial state of the pump field is in the  $l -$  photon Fock state,  $|l\rangle_0$ , and other anti-Stokes modes are in the vacuum states,  $|\varphi_{l,n}(0)\rangle$ . In general situation the initial bimodal field may be represented by the superposition of the populated states,  $|l_0\rangle_0 |l_1\rangle_1 \dots |l_n\rangle_n$ . Taking into consideration that the action of the operators,  $\hat{D}^+$ , and  $\hat{D}^-$ , on the collective state,  $\hat{D}^+ |j, m\rangle_r = \sqrt{(j + m + 1)(j - m)} |j, m + 1\rangle_r$ ,  $\hat{D}^- |j, m\rangle_r = \sqrt{(j + m)(j - m + 1)} |j, m - 1\rangle_r$ , we obtain the system of the  $N_r + 1$  equations for these operator—vectors,

$$\begin{aligned} \frac{d}{dt} \hat{X}_m(t) = & -i\delta m \hat{X}_m(t) - i\chi_n \sqrt{(j + m + 1)(j - m)} \hat{X}_{m+1}(t) \hat{\Lambda}_n^- \\ & - i\chi_n \sqrt{(j + m)(j - m + 1)} \hat{X}_{m-1}(t) \hat{\Lambda}_n^+, \\ & m = -j, -j + 1, \dots, j + 1, j. \end{aligned} \quad (19)$$

We observe that the system of equations (19) may be represented in the matrix form  $d\hat{X}/dt = \hat{X}\hat{D}$ , where  $\hat{X} = (\hat{X}_j, \hat{X}_{j-1}, \dots, \hat{X}_0, \dots, \hat{X}_{-j})$ . As follows from the description of the system (19), the principal determinant is described by matrix,  $\hat{D}$ , with non-commutative elements,  $\hat{D}_{km} = -i\delta_{k,m}m - i\chi_n\sqrt{(j+m+1)(j-m)}\hat{\Lambda}_n^-\delta_{k,m+1} - i\chi_n\sqrt{(j+m)(j-m+1)}\hat{\Lambda}_n^+\delta_{k,m-1}$ , so that is not so simple to find the eigenvectors of such a system of operator equations. In 5 we propose to modify the system of equation (19) introducing the modified operator-vectors in the system of equations described by a new principal matrix with commutative operator terms, so that the solution of this system of equation becomes possible for relative large number of atoms.

2. With an increasing number of atoms, we may solve the system of the equation (19), introducing the new operator-vectors, which depends on the atomic operators. This is possible if we know some commutative symmetries between the operators,  $\hat{\Lambda}_n^-$ , and  $\hat{\Lambda}_n^+$ , the action of which on the bimodal states,  $|\varphi_{l,n}(0)\rangle$ , may be regarded as annihilation or creation operators of the energy portion in the cavity field equal to,  $\hbar\omega \simeq \hbar\omega_r$ . Below we introduce for the special representation of these operators the  $su(2)$  and  $su(1, 1)$  symmetries in the multiple scattering processes. From proposed operator descriptions, we establish the commutation relations between the  $\hat{\Lambda}_n^-$  and  $\hat{\Lambda}_n^+$  operators.

Let us first introduce another representation of the solution for the Schrödinger equation (17),

$$|\psi(t)\rangle = \sum_{l_0, l_1, l_2, \dots, l_n} \beta_{l_0, l_1, l_2, \dots, l_n} \hat{F}_{l_0, l_1, l_2, \dots, l_n}(t) |\varphi_r(0)\rangle, \quad (20)$$

which can be used in the situation when the system of radiators a fixed in the cavity or evanescent zone of fiber, as this is represented in figure 2. As the pump photon flux passes through the cavity or fiber and enters into the multiple scattering processes with the localized ensemble of excited atoms we observe that in representation (20) the expressions,  $\hat{F}_{l_0, l_1, l_2, \dots, l_n} = \exp[-i\hat{H}_I t] |l_0\rangle_0 |l_1\rangle_1 \dots |l_n\rangle_n$ , are operator-vector of the cavity represented by the multiple scattered states of the bimodal field. As an interaction Hamiltonian acts only on these states, these vector operators depend on the atomic operators,  $\hat{D}^+$  and  $\hat{D}^-$ . The coefficients  $\beta_{l_0, l_1, l_2, \dots, l_n}$  represent the initial field superposition, in which was prepared photon pulse,  $|\varphi_{ph}(0)\rangle = \sum_{l_0, l_1, l_2, \dots, l_n} \beta_{l_0, l_1, l_2, \dots, l_n} |l_0\rangle_0 |l_1\rangle_1 \dots |l_n\rangle_n$ . The ensemble of atoms contains the same superposition of the Dicke states,  $|\varphi_r(0)\rangle = \sum_{m=-j}^j \alpha_m |j, m\rangle_r$ . This problem may also be simplified for relatively small number of photons and a finite number of atoms in the cavity. Below we reduce the system of equation evolution of system C to the well-known  $su(2)$  transition symmetry in the free and interaction parts of the Hamiltonian (1) which describes the multiple induced conversion of photons in the case of the big numbers of excited radiators. It is attractive from the physical point of view to describe this type of cooperative phenomenon in multiple Raman scattering for small number of scattering steps. The single-step cooperative Raman lasing was described in [21, 22, 36], the result of which can be obtained from the multistep scattering Hamiltonian (3) in the single step conversion,  $n = 1$ :  $\hat{\Lambda}_1^- = \hat{b}_1 \hat{c}_0^\dagger$ ,  $\hat{\Lambda}_1^+ = \hat{c}_0 \hat{c}_1^\dagger$ ;  $\hat{\Lambda}_1^z = (\hat{c}_1^\dagger \hat{c}_1 - \hat{c}_0^\dagger \hat{c}_0)/2$ . Here it is convenient to introduce the cooperative number,  $L = l/2$ , and angular momentum operators:  $L_{1/2}^- \equiv \hat{\Lambda}_1^-$ ,  $L_{1/2}^+ \equiv \hat{\Lambda}_1^+$  and  $L_{1/2}^z \equiv \hat{\Lambda}_1^z$ . For two steps of multiple scattering, we must take into consideration that,  $n = 2$ , adding the conversion Hamiltonian in the second mode,  $\hat{c}_2^\dagger (\hat{c}_2)$ , maybe reduced to two bimodal subsystems described by strength product,  $\hat{H}_I \sim \hat{D}^- \{k_1 \hat{E}_0^{(+)} \hat{E}_1^{(-)} + k_2 \hat{E}_1^{(+)} \hat{E}_2^{(-)}\}$ . Here, the generated anti-Stokes photons can be described in the language of negative and positive negative frequency strengths.

For example, in the first product  $\hat{E}_1^{(-)}(t, z) = q_1 \hat{c}_1^\dagger \exp(i\omega_1 t - ik_1 z)$  corresponds to generation of photons in the mode  $\omega_1 = \omega_0 + \omega_r$ , under the pump field,  $\hat{E}_1^{(+)} = q_0 \hat{c}_0 \exp(-i\omega_0 t + ik_0 z)$ . The positive frequency part of a new strength,  $\hat{E}_1^{(+)}(t, z) = q_1 \hat{c}_1 \exp(-i\omega_1 t + ik_1 z)$ , becomes the pump field for a next anti-Stokes photons from the negative frequency strength,  $\hat{E}_2^{(-)}(t, z) = q_2 \hat{c}_2^\dagger \exp(i\omega_2 t + ik_2 z)$ , in the interaction part of Hamiltonian,  $\hat{D}^- \hat{E}_1^{(+)} \hat{E}_2^{(-)}$ . According to figures 1 and 3 in the three level system the realization of  $su(2)$  commutation relations becomes possible when the scattering amplitudes,  $k_1 \sim 1/\Delta_1$  and  $k_2 \sim -1/\Delta_2$ , have same detunings from resonance,  $\Delta_1 \simeq -\Delta_2$  in the denominator. In this situation, we are able to introduce the new operators,  $\hat{\Lambda}_2^+ = (g_1 \hat{c}_0 \hat{c}_1^\dagger + g_2 \hat{c}_1 \hat{c}_2^\dagger)/\chi_2$ , and  $\hat{\Lambda}_2^- = (g_1 \hat{c}_1 \hat{c}_0^\dagger - g_2 \hat{c}_2 \hat{c}_1^\dagger)/\chi_2$ . In the two-steps Raman emission, the scattering amplitudes may be equal between them,  $|g_0| = |g_1|$ , so that the operators,  $\hat{\Lambda}_2^+$  and  $\hat{\Lambda}_2^-$ , belong to  $su(2)$  algebra. In this situation, introducing the renormalization coefficients,  $\chi_2 = g/\sqrt{2}$ , we may replaced the old operators by  $su(2)$  one,  $\hat{L}_1^- \equiv \hat{\Lambda}_2^+$ , and,  $\hat{L}_1^+ \equiv \hat{\Lambda}_2^-$ . We can introduce the operators,  $\hat{L}_1^z = \hat{c}_2^\dagger \hat{c}_2 - \hat{c}_0^\dagger \hat{c}_0$ ,  $\hat{L}_1^- = \sqrt{2} [\exp(-i\phi_1) \hat{c}_1 \hat{c}_0^\dagger \pm \exp(-i\phi_2) \hat{c}_2 \hat{c}_1^\dagger]$ ,  $\hat{L}_1^+ = \sqrt{2} [\exp(i\phi_1) \hat{c}_0 \hat{c}_1^\dagger \pm \exp(i\phi_2) \hat{c}_1 \hat{c}_2^\dagger]$ , which are described by the principal cooperative numbers,  $L = l$ , and the projection,  $m \equiv -l, \dots, l$ . Here the phases  $\phi_1$  and  $\phi_2$  take aleatory values and can be introduced in the new boson operators,  $\tilde{c}_1 = \exp(-i\phi_1) \hat{c}_1$  and  $\tilde{c}_2 = \exp(-i\phi_2) \hat{c}_2$ .

Similar results may be obtained for the three-step Raman emission,  $n = 3$ , in which we can introduce the operators,  $\hat{\Lambda}_3^+ = (g_1 \hat{c}_0 \hat{c}_1^\dagger + g_2 \hat{c}_1 \hat{c}_2^\dagger + g_3 \hat{c}_2 \hat{c}_3^\dagger) / \chi_3$ ;  $\hat{\Lambda}_3^- = (g_1 \hat{c}_1 \hat{c}_0^\dagger + g_2 \hat{c}_2 \hat{c}_1^\dagger + g_3 \hat{c}_3 \hat{c}_2^\dagger) / \chi_3$ . The special normalization of these operators choosing coefficients,  $g_1 / \chi_3 = g_3 / \chi_3 = \sqrt{3}$  and  $g_2 / \chi_3 = 2$ , may simplify the three steps of multiple Raman conversion, reducing this process to  $su(2)$  cooperative symmetry described by:  $\hat{L}_{3/2}^+ = \sqrt{3} \hat{c}_0 \hat{c}_1^\dagger + 2 \hat{c}_1 \hat{c}_2^\dagger + \sqrt{3} \hat{c}_2 \hat{c}_3^\dagger$ ;  $\hat{L}_{3/2}^- = \sqrt{3} \hat{c}_1 \hat{c}_0^\dagger + 2 \hat{c}_2 \hat{c}_1^\dagger + \sqrt{3} \hat{c}_3 \hat{c}_2^\dagger$ , and  $\hat{L}_{3/2}^z = -(3/2) \hat{c}_0^\dagger \hat{c}_0 - (1/2) \hat{c}_1^\dagger \hat{c}_1 + (1/2) \hat{c}_2^\dagger \hat{c}_2 + (3/2) \hat{c}_3^\dagger \hat{c}_3$ . In figures 1 and 3 the special detuning from resonance relative to the doublet of the excited virtual states opens the opportunity to realize the relations between these coefficients and experiment. In this case, the cooperative number becomes,  $L = 3l/2$ . The relation between the constant  $\chi_3$  and  $g_1$  is described by the expression,  $\chi_3 = g_1 / \sqrt{3}$ . As in the two steps of Raman conversion, the three steps of scattering are described by  $su(2)$  algebra,  $\hat{L}_s^+ \equiv \hat{\Lambda}_n^+$ ,  $\hat{L}_s^- \equiv \hat{\Lambda}_n^-$  and  $\hat{L}_s^z \equiv \hat{\Lambda}_n^z$ , in which we can represent the principal cooperative number,  $L$ , as a product of two numbers,  $L = sl$ . The first number,  $s = 3/2$ , is connected to the number of scattering steps,  $n = 3$ , and the second one with the number of quanta in the initial pump field,  $l$ . According to the mathematical induction, we can generalize this procedure for  $n$  - scattering steps described by angular momentum vectors,  $|L, m\rangle_f$  which are eigenvectors of  $\hat{L}_s^z$  operator,  $\hat{L}_s^z |L, m\rangle_f = m |L, m\rangle_f$ . Here the principal quantum number,  $L = ls$ , where  $s = n/2$ . The representation of these operators through the number of states becomes possible, introducing the steps parameter  $s = n/2$ ,

$$\begin{aligned}\hat{L}_s^+ &= \sum_{k=-s}^s \sqrt{(s+k)(s-k+1)} \hat{c}_{s+k-1} \hat{c}_{s+k}^\dagger; \\ \hat{L}_s^- &= \sum_{k=-s}^s \sqrt{(s+k)(s-k+1)} \hat{c}_{s+k-1}^\dagger \hat{c}_{s+k}; \\ \hat{L}_s^z &= \sum_{k=-s}^s k \hat{c}_{s+k}^\dagger \hat{c}_{s+k}.\end{aligned}\quad (21)$$

It is not difficult to control this relationship for single-, two-, and three- scattering steps described above. After the cooperative description (21), we return to unsymmetrical multiple conversions. We propose to construct the special states in which a great attention is given to the cooperative exchange process, with a portion of energy equal to,  $\hbar\omega \simeq \hbar\omega_r$ . This quasi-quanta can be generated or absorbed in the cavity bi-modes during the multiple scattering processes. In order to establish some simple relations between the conversion acts and the number of portions of energies that the cavity may obtain during the emission or absorption of converted photons. Below we reduce the interaction Hamiltonian (3), for this special value of the coefficients,  $g_1, g_1, \dots, g_n$  to  $su(2)$  symmetry. For this, we pass into the system of rotated coordinates with transition atomic frequency,  $\omega_r$ , and considering that the frequencies of generation photons, inside the cavity, are connected with a constant difference,  $\omega = \omega_{j+1} - \omega_j$ , by the multiple scattering expression,  $\omega_j = \omega_0 + j\omega$ , we represent the wave function  $|\psi(t)\rangle$  through the wave function in the rotation system of ordinate,  $|\bar{\psi}(t)\rangle$ , described by expression,  $|\psi(t)\rangle = \exp[it[\omega_r \hat{L}_s^z + \omega_r \hat{D}_2]] |\bar{\psi}(t)\rangle$ ,

$$\hat{H}_I = -\hbar\delta L_n^z + \hbar\chi_s \{\hat{L}_s^- \hat{D}^\dagger + \hat{D}^- \hat{L}_s^+\}, \quad (22)$$

in which the field operators and atomic one belong to the same symmetry, in which two independent Bloch vectors,  $L(L+1) = (\hat{L}_s^-)^2 + \{\hat{L}_s^- \hat{L}_n^\dagger + \hat{L}_s^- \hat{L}_s^+\} / 2$ , and,  $j(j+1) = \hat{D}_z^2 + \{\hat{D}^- \hat{D}^\dagger + \hat{D}^- \hat{D}^+\} / 2$ , are conserved. The first corresponds to the three cavity modes represented by the vector, in which the integer number  $L = sl$  depends on the number of photons of the pump field,  $l$ , and the number of the steps,  $n+1 = 2s$ , in the induced emission of multiple Raman scattering. The conversion operator of the photon between the Stokes and anti-Stokes modes takes place through the pump mode,  $L = 1/2, 1, 3/2, 2, \dots$ . For example, in the single step Raman scattering,  $L = l/2$ , and into two steps of multiple scattering processes  $L = (l/2) \times 2$ . To find the connection between the  $l$  and the number of pump photons  $n_0$ , below we represent the solution of Schrödinger equation (17) in another form than it was described in Exp.(18). Considering that the system of  $N_r$  excited atoms is prepared in the superposition of the states  $|j, m\rangle$  and the photon pulse in the superposition of  $su(2)$  states of the multiply scattered photons we represent the wave function (20) as a superposition sum of vector operators,

$$|\psi(t)\rangle = \sum_{p=0}^{2L} \beta_p \hat{F}_p(t) |\varphi_a(0)\rangle. \quad (23)$$

Here  $\hat{F}_p = \exp[-i\hat{H}_I t / \hbar] |L, -L+p\rangle_{ph}$ ,  $p = 0, 1, 2 \dots$  are the operator-vectors of multiple generated photons in the modes of cavity EMF,  $|\varphi_a(0)\rangle$  is the initial state of the atomic ensemble. According to this approach, these new vectors of the cavity field obey the system of differential equations,

$$\begin{aligned} \frac{d}{dt}\hat{F}_p(t) &= i\delta(p-L)\hat{F}_p(t) - i\chi_n\sqrt{p(2L-p+1)}\hat{F}_{p-1}(t)\hat{D}^+, \\ &\quad - i\chi_n\sqrt{(p+1)(2L-p)}\hat{F}_{p+1}(t)\hat{D}^-; \\ p &= 0, 1, \dots, 2l-1, 2L. \end{aligned} \quad (24)$$

This system of equations for field vectors,  $\{\hat{F}_p(t), p=0, \dots, 2L\}$ , have many analogies with the system of equations for the similar vectors of the atomic subsystem (19). Here we have introduced the specific interaction constants  $g_n, n=1, 2, \dots, n$ , which follows from the requirements of commutation relations for  $su(2)$  algebra (21). This procedure helps us to solve both systems (19) and (24) for a larger number of atoms or photons, respectively. The principal determinant of this system of equations also contains non-commutative elements and becomes difficult to apply the procedure of solutions of the system of equations with constant coefficients. In appendix we introduce a new vector, the system of differential equations, which is described by a principal determinant with commutative terms. The exact solutions of the system (24) are represented for principal quantum number  $L=3/2, 2$ .

To consecutively construct these states, let's consecutively act with operator  $\hat{\Lambda}_2^+$  on the initial two photon state in the pump mode,  $\hat{\Lambda}_2^+|2\rangle_0|0\rangle_1|0\rangle_2 = 2|1\rangle_0|1\rangle_1|0\rangle_2$ , which does not involve the absorption of the emitted photon. According to the next action of the excitation operator,  $\hat{\Lambda}_2^+$ , we obtain the superposition between the two cooperative processes connected with two quanta scattered in first anti-Stokes mode and cooperative re-conversion of the scattered photon,  $\hat{\Lambda}_2^+|1\rangle_0|1\rangle_1|0\rangle_2 = \sqrt{2}\{\sqrt{2}|0\rangle_0|2\rangle_1|0\rangle_2 + |1\rangle_0|0\rangle_1|1\rangle_2\}$ . Here, the first term describes the cooperative conversion of two pump photons into two scattered ones with frequency,  $\omega_1 = \omega_0 + \omega_r$ , while, the second term describes the reconverted of a new photon in the second scattered field with frequency  $\omega_2 = \omega_0 + 2\omega_r$ . The fourth-state represents the conversion of both photons: one in the first state and the other in the second scattered state in the double step of Raman conversion:  $\hat{\Lambda}_2^+\{\sqrt{2}|0\rangle_0|2\rangle_1|0\rangle_2 + |1\rangle_0|0\rangle_1|1\rangle_2\} = 3\sqrt{2}|0\rangle_0|1\rangle_1|1\rangle_2$ . The last term corresponds to the total conversion of all photons into the higher energy state,  $\hat{\Lambda}_2^+|0\rangle_0|1\rangle_1|1\rangle_2 = \sqrt{2}|0\rangle_0|0\rangle_1|2\rangle_2$ . Begging with  $L \geq 2$ , such a superposition plays an important role in multiple scattering processes.

If we consider that the coefficients  $g_p$  increase proportional to the step number of multiple Raman transitions,  $p$ , another type of symmetry follows from the representation of commutation relations (A1) and (A2). Indeed, for a big number of multiple scattering processes,  $n \rightarrow \infty$ , coefficients,  $g_p = g_0p$ , and,  $\chi_n = g_0$ , the operators

$$\hat{\Lambda}^- = \lim_{n \rightarrow \infty} \sum_{k=0}^{n-1} (k+1)\hat{c}_{k+1}\hat{c}_k^\dagger; \quad \hat{\Lambda}^+ = \lim_{n \rightarrow \infty} \sum_{k=0}^{n-1} (k+1)\hat{c}_k\hat{c}_{k+1}^\dagger, \quad (25)$$

and

$$\hat{\Lambda}_z = \frac{1}{2} \lim_{n \rightarrow \infty} \sum_{k=0}^{n-1} (k+1)^2 \{\hat{c}_{k+1}\hat{c}_{k+1}^\dagger - \hat{c}_k\hat{c}_k^\dagger\},$$

belong to  $su(1, 1)$  symmetry with eigenvectors,  $|\kappa, \kappa+n\rangle$ , of operator  $\hat{\Lambda}_z, \hat{\Lambda}_z|\kappa, \kappa+p\rangle = (\kappa+p)|\kappa, \kappa+p\rangle$  in which the defined operators excited and lowering (25)  $\hat{\Lambda}^+$ , and  $\hat{\Lambda}^-$ , with actions  $\hat{\Lambda}^+|\kappa, \kappa+p\rangle = \sqrt{(2\kappa+p)(p+1)}|\kappa, \kappa+p+1\rangle, \hat{\Lambda}^-|\kappa, \kappa+p\rangle = \sqrt{(2\kappa+p-1)p}|\kappa, \kappa+p-1\rangle, p=0, 1, 2, \dots, r, \dots$ . It is not complicated to demonstrate that this operators belong to this symmetry,

$$\begin{aligned} [\hat{\Lambda}^+, \hat{\Lambda}^-] &= \lim_{n \rightarrow \infty} \sum_{k=0}^{n-1} (k+1)^2 \{\hat{c}_{j+1}\hat{c}_{k+1}^\dagger - \hat{c}_k\hat{c}_k^\dagger\} = 2\hat{\Lambda}_z; \quad 0 \\ [\hat{\Lambda}^+, \hat{\Lambda}_z] &= \lim_{n \rightarrow \infty} \frac{1}{2} \sum_{k=0}^{n-1} \{-2(k+1)^2 + k^2 + (k+2)^2\theta(n-k-2+0.5)\} \\ &\quad \times (k+1)\hat{c}_k\hat{c}_{k+1}^\dagger = \hat{\Lambda}^+. \end{aligned} \quad (26)$$

Here in the process of system excitation of cavity field in multiple Raman process is conserved in the vector,  $\kappa(\kappa-1) = \hat{\Lambda}_z^2 - \{\hat{\Lambda}^-\hat{\Lambda}^+ + \hat{\Lambda}^+\hat{\Lambda}^-\}/2$ . From the definition of operators,  $\hat{\Lambda}_z, \hat{\Lambda}^-$  and  $\hat{\Lambda}^+$ , and initially prepared. We observed that multiple scattering effects are manifested for a large number of photons. To observe the superposition of multiple scattering photons in a large number of modes, it is necessary to have a large ensemble of exciting atoms that may multiply scattered under the action of the pump field. Following up from

the estimation of Exp. (36), for four atoms the possibility to observe the high mode correlations,  $E_3^2$ , becomes impossible due to the small number of scattering radiators in the system. There exist two possibilities. The first action is to return to the Schrödinger equation for a large ensemble of atoms. The second possibility is to pump the excited atoms into the resonator and obtain the lasing effect in multiple scattering process. The first possibility is limited by the possibilities to solve exactly the system of operator vectors for the large number of atoms or large numbers of photons in the pump field. Such a problem involves a large degree of freedom in the proposed approach, which limits the possibility to represent in analytic form the solutions of the Schrödinger equation (see appendix). The second approach is connected to finding the system of wave functions like,  $(\hat{A}_n^+)^k |1\rangle_0 |0\rangle_{1\dots n}$ , of multiple scattered fields on the Hilbert space. In this space, we may construct the evolution of such quantum correlations of cavity excitations during the multiple scattered lasing. From the initial state of the cavity field,  $|\varphi_{ph}(0)\rangle = |l\rangle_0 |0\rangle_{1\dots n}$ , follows that the coefficient,  $\kappa = l/2$ .

Let us find the solution of the Schrödinger equation (17) in the form represented by expressions,  $|\psi_{j,\kappa}(t)\rangle = \sum_{p=0}^{\infty} \beta_p \hat{\Phi}_{\kappa+p}(t) |\psi_j(0)\rangle$ , where the expressions,  $\{\hat{\Phi}_{\kappa+p} = \exp[-i\hat{H}_I t/\hbar] |\kappa, \kappa+p\rangle_{ph}\}$ ,  $p = 0, 1, 2, \dots$ , are an infinite set of vector-operators obtained in the multiple generated photons in the modes of the cavity EMF. According to the  $su(1, 1)$  algebra roles, the new system of equations follows from the Schrödinger equation (17)

$$\begin{aligned} \frac{d}{dt} \hat{\Phi}_{\kappa+p}(t) &= i\delta(p + \kappa) \hat{\Phi}_p(t) - i\chi \sqrt{(2\kappa + p - 1)p} \hat{\Phi}_{\kappa+p-1}(t) \hat{D}^+ \\ &\quad - i\chi \sqrt{(2\kappa + p - 1)p} \hat{\Phi}_{\kappa+p+1}(t) \hat{D}^-; \\ p &= 0, 1, 2, \dots, k, \dots, \end{aligned} \quad (27)$$

where and  $\chi = g_1$ . The system of equation (A15) with commutative terms of principal determinants may be obtained from (27) by the substitution,  $\hat{\Upsilon}_p(t) = \hat{\Phi}_{\kappa+p}(t) (\hat{D}^-)^p$ , and applying the commutation relations (26).

We emphasize that there exist two possibilities to solve exactly the Schrödinger equation (17). The first method consists of the exact solution of  $2j + 1$  operator vectors declared in the system of equations (19) or (A4). This method is applicable for a relatively small number of radiators in comparison with the number of photons in the closed system. The second approach is connected to the possibilities of the application of symmetries  $su(2)$  and  $su(1, 1)$  in the multiply scattering process described by the system of equations (A.7) and (27) for relative small number of photons in the field. In the case of the same small number of photons and atoms, these two methods must coincide. We observe that, at first glance, for a few atoms, the number of equations in the system (27) is infinite. But maybe not so, because the chain of equations for the field vectors  $\hat{\Upsilon}_p(t) = \hat{\Phi}_{\kappa+p}(t) (\hat{D}^-)^p$ , is truncated when the number of atoms  $N_a < p$  (in this situation  $(\hat{D}^-)^p = 0$ ).

### 3. Losses of excitations in the cooperative multiple scattering

Let us estimate the generation of converted photons in the free space. In this approach, we can use in the right-hand side of the above equations the solutions from the appendix. The possible multiple scattering effect in the subsystem of two excited atoms in interaction with the cavity field can be obtained if we consider that initially the field is prepared in the pump mode  $|l\rangle_0, |0\rangle_1, \dots, |0\rangle_n$ . Neglecting the detuning,  $\delta = 0$ , and vacuum re-normalization, the simple solution for atomic vector operators (A3) we obtain the following solution,

$$\begin{aligned} |\psi_l^r(t)\rangle &= \left\{ 1 - 4g_1^2 l \frac{\sin^2[\Omega_{l1}t/2]}{\Omega_{12}^2} \right\} |1, 1\rangle_r \otimes |l\rangle_0, |0\rangle_1, \dots, |0\rangle_n \\ &\quad - ig_1 \sqrt{2l} \frac{\sin[\Omega_{l1}t]}{\Omega_{l1}} |l-1\rangle_0 |1\rangle_1 \dots |0\rangle_n \otimes |1, 0\rangle_r \\ &\quad - 4g_1 \sqrt{l} \frac{\sin^2[\Omega_{l1}t/2]}{\Omega_{l1}^2} \{g_1 \sqrt{2(l-1)} |l-2\rangle_0, |2\rangle_1 |0\rangle_2 \dots |0\rangle_n \\ &\quad + g_2 |l-1\rangle_0, |0\rangle_1 |1\rangle_2 \dots |0\rangle_n\} \otimes |1, -1\rangle_r, \end{aligned} \quad (28)$$

where  $\Omega_{l1} = \sqrt{2(3g_1^2 l - 2g_1^2 + g_2^2)}$ . As follows from the definition of Rabi frequency,  $\Omega_{12}$ , and from the last term of wave function (28) the migration of the photon in the next step of multiple scattering process,  $g_2 \hat{c}_2^\dagger \hat{c}_1 \hat{D}^-$ , is possible to. Introduce the solution ((28)) into the loss rate of cvasi-particles from cavity described by Exp. (11), we observe this migration in non-adjacent modes described by the cooperative rate  $\Gamma_{0,2}$ ,



$$\begin{aligned}
\frac{d\langle\hat{W}_C(t)\rangle}{dt} = & -4\Gamma_0^{(1)}l\left\{2g_1^2l\frac{\cos[\Omega_{1l}t]-1}{\Omega_{1l}^2}+1\right\}^2 \\
& -2^3g_1^2l\frac{\sin^2[\Omega_{1l}t]}{\Omega_{12}^2}\{\Gamma_0^{(1)}(l-1)+2\Gamma_0^{(1)}\} \\
& -2^4g_1g_2l\Gamma_{0,2}\frac{\cos[\Omega_{1l}t]-1}{\Omega_{1l}^2}\left\{2g_1^2l\frac{\cos[\Omega_{1l}t]-1}{\Omega_{1l}^2}+1\right\} \\
& -2^3g_2g_1l\Gamma_2^{(2)}\frac{[\cos[\Omega_{1l}t]-1]^2}{\Omega_{1l}^4}.
\end{aligned} \tag{29}$$

According to Exp. (29) for a large number of pump photons, we may neglect the rate,  $l\Gamma_p^{(2)}$  in comparison with  $l^2\Gamma_p^{(1)}$  because the induced scattering from the ground state is small due to the large values of number of photons,  $l$ . But at this stage, the contribution of the correlations of the non-adjacent modes in the losses of the quasi—particles from the ‘cavity+field’ system described by the coefficients,  $\Gamma_{0,2}$ .

With increasing the number of excited atoms in the system  $2j$ , the possibilities of multiple Raman scattering increases too. According to the system of equations (A7) we obtain a similar wave function like (29) in which the quantum number,  $L = l/2$  for photon excitations is substituted by number  $j$ . We may study this effect using the exact solutions for small values of the quantum number  $L = sl = 1$  described by the vectors (A10). In this situation we have two possibilities. First scattering process corresponds to the single step and two photon in the pump and scattered modes described by the states:  $|2\rangle_0|0\rangle_1$ ;  $|1\rangle_0|1\rangle_1$ , and  $|0\rangle_0|2\rangle_1$ . Second one correspond to one photon ( $l = 1$ ) in two—step scattering ( $s = 1$ ), described by the states:  $|1\rangle_0|0\rangle_1|0\rangle_2$ ;  $|0\rangle_0|1\rangle_1|0\rangle_2$ , and  $|0\rangle_0|0\rangle_1|1\rangle_2$ . The first case have similarities with solution (28) in which the quantum number,  $L = l/2$  for photon excitation is substituted by number  $j$ . The single photon in the two-step scattering conversion by arbitrary number of atoms, have some peculiarities in the lossesbehavior. In this case the solution of Schrödinger equation obtained from initial state,  $|j, j\rangle_a \otimes |1\rangle_0|0\rangle_1|0\rangle_2$ , is described by the expression,

$$\begin{aligned}
|\psi_j^f(t)\rangle = & \left\{1 - 4jg^2\frac{\sin^2(\Omega_{1j}t/2)}{\Omega_{1j}^2}\right\}|j, j\rangle_r \otimes |1\rangle_0|0\rangle_1|0\rangle_2 \\
& - ig\sqrt{2j}\frac{\sin(\Omega_{1j}t)}{\Omega_{1j}}|0\rangle_0|1\rangle_1|0\rangle_2|j, j-1\rangle_r \\
& - 4g^2\sqrt{j(2j-1)}\frac{\sin^2(\Omega_{1j}t/2)}{\Omega_{1j}^2}|0\rangle_0|0\rangle_1|1\rangle_2|j, j-2\rangle_r,
\end{aligned}$$

where  $\Omega_{1j} = g\sqrt{(3j-1)/2}$ . According to this solution, the loss rate of of cvasi particles from cavity (11) takes another form,

$$\begin{aligned}
\frac{d\langle\hat{W}_C(t)\rangle}{dt} = & -4j\Gamma_0^{(1)}\left\{1 + 2jg^2\frac{[\cos(\Omega_{1j}t)-1]}{\Omega_{1j}^2}\right\}^2 \\
& - 2^4jg^2(2j-1)\frac{\sin^2(\Omega_{1j}t)}{\Omega_{1j}^2}\Gamma_1^{(1)} \\
& - 2^4\left\{1 + 2jg^2\frac{[\cos(\Omega_{1j}t)-1]}{\Omega_{1j}^2}\right\}\sqrt{j(2j-1)}\frac{[\cos(\Omega_{1j}t)-1]}{\Omega_{1j}^2}\Gamma_{0,2} \\
& - 2^4g^4j(2j-1)^2\frac{[\cos(\Omega_{1j}t)-1]^2}{\Omega_{1j}^4}\Gamma_2^{(2)}
\end{aligned} \tag{30}$$

Here, we observe an involvement of the correlation between the photons from non-adjacent modes of first and second scattered steps. But it is not so large superposition,  $\sim\Gamma_{0,2}$ , in comparison with first term proportional,  $j\Gamma_0^{(1)}$ . Due to the small number of photon and large cooperative effects between the atoms the probability of scattered photon in the external field by atoms from ground states increases too,  $\sim j\Gamma_2^{(2)}$ .

For the large numbers  $l$  and  $s$  this process involve the superposition between the converted photon states belonging to non-adjacent modes through the excited states of atomic subsystem. This effect is studied in in muti-step Raman lasing of [37]. From section 2 follows that such superposition appears for two quanta,  $l = 2$  in the two steps Raman conversion,  $s = 1$ , in which is realized the cooperative state,  $|2, 0\rangle_f$ . In this situation, it is better to study this simple superposition using the solution (A11) for the set of five field vectors, which describes the entanglement between photons belonging to scattering components using the second moments (14) and normal fluctuations (15). The wave function for two quanta in the two steps scattering process can be obtained from the action of the vector (A11), on the exited system of atoms  $|j, j\rangle$ ,



$$|\psi^f(t)\rangle = [\hat{A}_0 + \hat{A}_1 \cos(\hat{\Omega}_1 t) + \hat{A}_2 \cos(\hat{\Omega}_2 t) + \hat{B}_1 \sin(\hat{\Omega}_1 t) + \hat{B}_2 \sin(\hat{\Omega}_2 t)] |j, j\rangle, \quad (31)$$

where the coefficients  $\hat{A}_0$ ,  $\hat{A}_i$  and  $\hat{B}_i$ ,  $i=1,2$ , are defined by the expressions (A12), (A13) and (A14). Taking into consideration the inequality,  $|\chi_{p,k}|^2 \ll |g_p|^2$ , on the right side of the expressions (11) and (14), we can approximate the density matrix of the closed system by the expression,  $\hat{\rho}(t) \approx |\psi(t)\rangle \langle \psi(t)|$ . As processes connected with scattering instead of coefficients  $\Gamma_p^{(1)}$  for simplicity of the expressions for higher moment we introduce  $\Gamma_p$ . In this approximation, we can calculate the correlates,

$$\langle \hat{c}_p^\dagger(t) \hat{c}_p(t) \hat{D}^+(t) \hat{D}^-(t) \rangle \approx \langle \psi(t) | \hat{c}_p^\dagger \hat{c}_p \hat{D}^+ \hat{D}^- | \psi(t) \rangle,$$

and

$$\begin{aligned} & \langle \hat{c}_p^\dagger(t) \hat{c}_{p_1}^\dagger(t) \hat{c}_{p'}(t) \hat{c}_{p_1}(t) \hat{D}^+(t) \hat{D}^+(t) \hat{D}^-(t) \hat{D}^-(t) \rangle \\ & = \langle \psi^f(t) | \check{c}_p^\dagger(t) \check{c}_{p_1}^\dagger(t) \check{c}_{p'}(t) \check{c}_{p_1}(t) \check{D}^+(t) \check{D}^+(t) \check{D}^-(t) \check{D}^-(t) | \psi^f(t) \rangle. \end{aligned} \quad (32)$$

According to the Schrödinger equation (17), the operators must be in the interaction picture,  $\hat{c}_p^\dagger(t) = \hat{c}_p^\dagger(0) \exp[i\omega t]$ ,  $\hat{D}^+(t) = \hat{D}^+(0) \exp[i\omega t]$ . As follows from the correlations (32) contains the photons scattered from different steps of multiple Raman emissions. For example if in the solution for principal quantum number,  $L = 2$ , we have the superposition  $|\psi(t)\rangle \sim \sqrt{2} |0\rangle_0 |2\rangle_1 |0\rangle_2 + |1\rangle_0 |0\rangle_1 |1\rangle_2$ . In this situation we may create the vacuum state acting with  $\check{c}_1(t) \check{c}_1(t)$  on the first term,  $\check{c}_1^2 |0\rangle_0 |2\rangle_1 |0\rangle_2 = \sqrt{2} |0\rangle_0 |0\rangle_1 |0\rangle_2$  and another way is obtained by acting with operators  $\check{c}_0(t) \check{c}_2(t)$  on the second function from superposition,  $\check{c}_2 \check{c}_0 |1\rangle_0 |0\rangle_1 |1\rangle_2 = |0\rangle_0 |0\rangle_1 |0\rangle_2$ . The non-oscillatory part of the photon correlates will be proportional to the sum of three types of correlates  $\langle \check{c}_p^\dagger(t) \check{c}_{p_1}^\dagger(t) \check{c}_{p'}(t) \check{c}_{p_1}(t) \rangle \sim \langle \check{c}_1^2(0) \check{c}_0(0) \check{c}_2(0) \rangle + \langle \check{c}_0^2(0) \check{c}_2^2(0) \check{c}_1^2(0) \rangle + \langle \check{c}_0^2(0) \check{c}_2^2(0) \check{c}_0(0) \check{c}_2(0) \rangle$ . First and second correlations contain the entanglement between the two photons from pump and first scattered mode and the photons from pump and second step scattered one, the third correlation contains only the photons from the same mode.

The algorithm of this procedure of factorization of the correlates like,  $\langle (\hat{\Lambda}_n^-)^m \check{c}_p^\dagger(t) \check{c}_{p_1}^\dagger(t) \check{c}_{p'}(t) \check{c}_{p_1}(t) (\hat{\Lambda}_n^+)^m \rangle$ , is not so well established for the large number of photons and large number of steps in Raman scattering. For this, we return to System of equation (A4) and construct the solution for four atoms involved in the multiple Raman conversion. Considering four radiators prepared in the excited state, we can obtain the wave function using the solution of the system of equations (A4) for atomic vector operators,  $\{\hat{R}_m(t), m = -2, \dots, 2\}$ . Acting with the vector operator

$$\hat{R}_j(t) = \hat{Y}_0 + \hat{Y}_1 \cos(\hat{\Omega}_{r1} t) + \hat{Y}_2 \cos(\hat{\Omega}_{r2} t) + \hat{X}_1 \sin(\hat{\Omega}_{r1} t) + \hat{X}_2 \sin(\hat{\Omega}_{r2} t),$$

on the initial state of cavity field,  $|L, -L\rangle_f$  in the analogy with the solution (31) we obtain the wave function,

$$|\psi^r(t)\rangle = [\hat{Y}_0 + \hat{Y}_1 \cos(\hat{\Omega}_{r1} t) + \hat{Y}_2 \cos(\hat{\Omega}_{r2} t) + \hat{X}_1 \sin(\hat{\Omega}_{r1} t) + \hat{X}_2 \sin(\hat{\Omega}_{r2} t)] |L, -L\rangle_f,$$

in which the small number of photons and large number of atoms used analytic representation of the solution (31) is replaced by relative small number of radiators,  $N_r = 4$ , and big numbers of photons with possibilities to be converted into large number of scattering steps. Here the coefficients are

$$\begin{aligned} \hat{Y}_0 & = |2, 2\rangle_r - 8\chi_n^4 |2, 2\rangle_r \Delta_1^{su} (2\Delta_4^{su} + 3\Delta_3^{su}) \\ & \quad + \sqrt{6} |2, 0\rangle_r (\hat{\Lambda}_n^+)^2 \Delta_4^{su} - 3 |2, -2\rangle_r (\hat{\Lambda}_n^+)^4 \hat{\Omega}_{r2}^{-2} \hat{\Omega}_{r1}^{-2}; \end{aligned} \quad (33)$$

$$\begin{aligned} \hat{Y}_i & = 2\chi_n^2 \{2 |2, 2\rangle_r \Delta_1^{su} [2\chi_n^2 (3\hat{\Delta}_3^{su} + 2\hat{\Delta}_4^{su}) - \hat{\Omega}_{ri}^2] \\ & \quad + \sqrt{6} |2, 0\rangle_r (\hat{\Lambda}_n^+)^2 [4\chi_n^2 \hat{\Delta}_4^{su} - \hat{\Omega}_{ri}^2] \\ & \quad - 3 \times 4\chi_n^2 |2, 2\rangle_r (\hat{\Lambda}_n^+)^4 \frac{1}{\hat{\Omega}_i^2 (\hat{\Omega}_j^2 - \hat{\Omega}_i^2)}; i, j = 1, 2; i \neq j; \end{aligned} \quad (34)$$

$$\begin{aligned} \hat{X}_i & = 2i\chi_n \{ |2, 1\rangle_r \hat{\Lambda}_n^+ [\hat{\Omega}_{ri}^2 - 2\chi_n^2 (3\hat{\Delta}_3^{su} + 2\hat{\Delta}_4^{su})] \\ & \quad + 3 \times 2\chi_n^2 |2, -1\rangle_r (\hat{\Lambda}_n^+)^3 \frac{1}{\hat{\Omega}_i (\hat{\Omega}_j^2 - \hat{\Omega}_i^2)}; i, j = 1, 2; i \neq j. \end{aligned} \quad (35)$$

These cooperative phenomena between the conversion of the photon in the multiple reabsorbing of the quanta on the next conversion steps can be observed using the higher moments of the energy, losses  $\langle (d\hat{W}_e/dt)^\alpha \rangle$ ,  $\alpha = 1, 2, \dots$ . As follows from the coefficients (33), (34), (35) and (21), the maximum of conversion rate in the next steps may be described by the correlation function,  $\langle (\hat{\Lambda}_n^-)^4 \check{c}_p^\dagger(t) \check{c}_{p_1}^\dagger(t) \check{c}_{p'}(t) \check{c}_{p_1}(t) (\hat{\Lambda}_n^+)^4 \rangle$ , obtained from expressions for  $\hat{Y}_i$ . But this terms doesn't give the

contribution in the higher order moments,  $\langle : (d\hat{W}_e/dt)^\alpha : \rangle$ , due to the fact that we used the exact solution of Schrödinger equation for small number of atoms involved in the scattering processes,  $N_r = 4$ . In such situation the mean value of atomic correlations becomes equal to zero in correlation function,  $\langle 2, -2 | (\hat{D}^+)^\alpha (\hat{D}^-)^\alpha | 2, m \rangle_r = 0$ , for  $|m - \alpha| > 2$ . We can estimate higher order moments of the cavity excitations,

$$\begin{aligned} \left\langle : \left( \frac{d}{dt} \hat{W}_e(t) \right)^\alpha : \right\rangle &= \sum_{p, p'} \dots \sum_{p_\alpha, p'_\alpha} \Gamma_{p_1, p'_1} (p'_1 + 1) \dots \Gamma_{p_\alpha, p'_\alpha} (p'_\alpha + 1) \\ &\times \{ C_{S1}(t) \langle L, -L | \check{c}_{p_1}^\dagger(t) \dots \check{c}_{p_{\alpha 2}}^\dagger(t) \check{c}_{p'_1}(t) \dots \check{c}_{p'_{\alpha}}(t) | L, -L \rangle_f K_2^\alpha \\ &+ S_{N1}(t) \langle L, -L | \hat{\Lambda}_n^- \check{c}_{p_1}^\dagger(t) \dots \check{c}_{p_{\alpha 2}}^\dagger(t) \check{c}_{p'_1}(t) \dots \check{c}_{p'_{\alpha}}(t) \hat{\Lambda}_n^+ | L, -L \rangle_f K_1^\alpha \\ &+ C_{S2}(t) \langle L, -L | (\hat{\Lambda}_n^-)^2 \check{c}_{p_1}^\dagger(t) \dots \check{c}_{p_{\alpha 2}}^\dagger(t) \check{c}_{p'_1}(t) \dots \check{c}_{p'_{\alpha}}(t) (\hat{\Lambda}_n^+)^2 | L, -L \rangle_f K_0 \\ &+ S_{N2}(t) \langle L, -L | (\hat{\Lambda}_n^-)^3 \check{c}_{p_1}^\dagger(t) \dots \check{c}_{p_{\alpha 2}}^\dagger(t) \check{c}_{p'_1}(t) \dots \check{c}_{p'_{\alpha}}(t) (\hat{\Lambda}_n^+)^3 | L, -L \rangle_f K_{-1} \\ &+ [(p_i, p'_i) \rightarrow (p'_i, p_i), i = 1, \dots, \alpha] \}. \end{aligned} \quad (36)$$

Here  $[(p_i, p'_i) \rightarrow (p'_i, p_i), i = 1, \dots, \alpha]$  represents the sum of terms with the possible permutations of indexes  $p_i$  with  $p'_i$ . The atomic correlations  $K_m^\alpha = \langle 2, m | (\hat{D}^+)^\alpha (\hat{D}^-)^\alpha | 2, m \rangle_r$ , can be easily estimated,  $\langle 2, m | (\hat{D}^+)^\alpha (\hat{D}^-)^\alpha | 2, m \rangle_r = (2 + m)!(2 - m + \alpha)! / [(2 + m - \alpha)!(2 - m)!]$ . For the losses of cavity excitations,  $\langle d\hat{W}_e(t)/dt \rangle$ , we have the correlations:  $K_2^1 = 4$ ;  $K_1^1 = 6$ ;  $K_0^1 = 6$ ;  $K_{-1}^1 = 6$ . For square losses,  $\langle : (d\hat{W}_e(t)/dt)^2 : \rangle$ , we have the coefficients:  $K_2^2 = 24$ ;  $K_1^2 = 36$ ;  $K_0^2 = 24$ ;  $K_{-1}^2 = 0$ . The time dependent coefficients of expression (36) are represented through the Rabi oscillation expressions,

$$\begin{aligned} C_{S1}(t) &= \left\{ 1 + \frac{32L\chi_n^4}{\Omega_1^2\Omega_2^2(\Omega_1^2 - \Omega_2^2)} [\Omega_1^2(\cos(\Omega_2 t) - 1) \right. \\ &\quad \left. - \Omega_2^2(\cos(\Omega_1 t) - 1)](17L - 21) + \frac{8L\chi_n^2}{\Omega_1^2 - \Omega_2^2} [\cos(\Omega_1 t) - \cos(\Omega_2 t)] \right\}^2; \\ C_{S2}(t) &= \left\{ \frac{32\chi_n^4(3 - 2L)}{\Omega_2^2\Omega_1^2(\Omega_1^2 - \Omega_2^2)} [\Omega_1^2(\cos(\Omega_2 t) - 1) - \Omega_2^2(\cos(\Omega_1 t) - 1)] \right. \\ &\quad \left. + \frac{2\chi_n^2}{(\Omega_1^2 - \Omega_2^2)} [\cos(\Omega_1 t) - \cos(\Omega_2 t)] \right\}^2; \\ S_{m1}(t) &= \frac{1}{(\Omega_1^2 - \Omega_2^2)^2} \{ \Omega_2 \sin(\Omega_2 t) - \Omega_1 \sin(\Omega_1 t) \\ &\quad - \frac{4\chi_n^2(17L - 21)}{\Omega_1\Omega_2} [\Omega_1 \sin(\Omega_2 t) - \Omega_2 \sin(\Omega_1 t)] \}^2; \\ S_{m2}(t) &= \frac{3^2 \times 2^4 \chi_n^6}{\Omega_2^2\Omega_1^2(\Omega_1^2 - \Omega_2^2)^2} \{ \Omega_1 \sin(\Omega_2 t) - \Omega_2 \sin(\Omega_1 t) \}^2. \end{aligned}$$

Here the Rabi frequencies were calculated on the cavity field state  $||L, -L\rangle_f$  in the normal representation of the moments (36),  $\Omega_{1,2} = \chi_n(50L - 48) \pm 2\sqrt{3}\chi_n\sqrt{99L^2 - 16L + 114}$ , where sign, '+' was chosen for  $\Omega_1$ , and sign '-', is for  $\Omega_2$ . Taking into consideration that the initial state of the field is expressed through the multiple scattering modes,  $|L, -L\rangle_f = |l\rangle_f |0\rangle_1 \dots |0\rangle_m$ , we can easily estimate first correlation,  $E_1^r = \langle L, -L | \check{c}_{p'_1}^\dagger(t) \check{c}_{p_1}^\dagger(t) \dots \check{c}_{p'_r}^\dagger(t) \check{c}_{p_r}(t) \check{c}_{p_1}(t) \dots \check{c}_{p_r}(t) | L, -L \rangle_f = l(l-1)\dots(l-r+1)\delta_{p_1,0}\delta_{p_1',0} \dots \delta_{p_r,0}\delta_{p_r',0}$ . It is not simple to calculate second correlates from the expression (36) after the action of operators  $(\hat{\Lambda}_n^-)^k$  and  $(\hat{\Lambda}_n^+)^k$ , on the bra- and ket-vectors of expression  $E_k^r \sim \langle L, -L + k | \check{c}_{p'_1}^\dagger(t) \dots \check{c}_{p'_r}^\dagger(t) \check{c}_{p_1}(t) \dots \check{c}_{p_r}(t) | L, -L + k \rangle_f$ . At first glance, we observe some de-correlations in the analogy with Wick theorem proposed in quantum statistical mechanics (see for example [38]), but the particular examples like,  $\langle 2, 0 | \check{c}_1^{\dagger 2}(t) \check{c}_0(t) \check{c}_2(t) | 2, 0 \rangle$ , described by Exp. (32), demonstrate that the index pairing between the creation and annihilation operators,  $\check{c}_{p'_i}^\dagger(t)$ ,  $\check{c}_{p_i}^\dagger(t)$ , and,  $\check{c}_p(t) \check{c}_{p_i}(t)$ , don't contain all non-zero terms. In order to calculate the above correlations, we propose to permute the annihilation (generation) operators from left-hand (right-hand) to right-hand (left-hand) one on the correlation function like,  $E_k^r = \langle L, -L | \check{c}_{p_1}^\dagger(t) \dots \check{c}_{p_r}^\dagger(t) \check{c}_{p_1}(t) \dots \check{c}_{p_r}(t) (\hat{\Lambda}_n^+)^k | L, -L \rangle_f$ . For this we must commute the annihilation operators,  $\check{c}_{p_1}(t)$ ,  $c_{p_2}$  and  $\check{c}_{p_3}(t)$ , with representation of excited operators,  $\hat{\Lambda}_n^+ \equiv \hat{L}_n^+$ , through  $c_p^\dagger$  and  $\check{c}_p(t)$  defined in Exps. (21). Doing this, we can act with the set of operators,  $\check{c}_{p_1}(t)$ ,  $c_{p_2} \dots \check{c}_{p_r}(t)$ , on the initial state,  $\check{c}_{p_1}(t)$ ,  $c_{p_2} \dots \check{c}_{p_r}(t) | L, -L \rangle_f = \delta_{p_1,0} \delta_{p_2,0} \dots \delta_{p_r,0} \sqrt{l(l-1)\dots(l-r+1)} |l-2\rangle_f |0\rangle_1 \dots |0\rangle_n \exp[-i(\omega_{p_1} + \omega_{p_2} + \dots + \omega_{p_r})t]$ . The simple transfer to the right-hand side into correlation

function of operator,  $\hat{c}_p$ , through the  $su(2)$ -operator,  $L_n^+$ , becomes,  $\hat{c}_p \hat{L}_n^+ = \sqrt{p(2s-p+1)} \hat{c}_{p-1} + \hat{L}_n^+ \hat{c}_p$ . As we are interested in the square losses, we take as an example the correlation,  $E_2^2$ , but the method can be extended to other correlations which contains higher order of operators,  $(\hat{L}_n^+)^k$ ,  $k = 1, 2, 3, \dots$ . We give below only the transfer of one  $\hat{c}_{p_2}$  operator through the  $(L_n^+)^2$ ,

$$\begin{aligned} \hat{c}_{p_2} (\hat{L}_n^+)^2 &= (\hat{L}_n^+)^2 \hat{c}_{p_2} + 2\hat{L}_n^+ \sqrt{p_2(2s-p_2+1)} \hat{c}_{p_2-1} \\ &+ \sqrt{p_2(p_2-1)(2s-p_2+1)(2s-p_2+2)} \hat{c}_{p_2-2}. \end{aligned}$$

Doing the similar transfer with operator,  $\hat{c}_{p_1}$ , we obtain the amplitude,

$$\begin{aligned} \hat{c}_{p_1} \hat{c}_{p_2} (\hat{L}_n^+)^2 |L, -L\rangle_f &= \sqrt{l(l-1)} \{ (\hat{L}_n^+)^2 \delta_{p_2,0} \delta_{p_1,0} \\ &+ 2\sqrt{2s} \hat{L}_n^+ [\delta_{p_2,0} \delta_{p_1-1,0} + \delta_{p_1,0} \delta_{p_2-1,0}] \\ &+ [\sqrt{2!2s(2s-1)} [\delta_{p_1-2,0} \delta_{p_2,0} + \delta_{p_2-2,0} \delta_{p_1,0}] \\ &+ 2s \delta_{p_2-1,0} \delta_{p_1-1,0}] |L_2, -L_2\rangle_f. \end{aligned} \quad (37)$$

Here we observe the possible interference of the amplitudes of pump field with frequency  $\omega = \omega_0$  (see  $p = 0$ ) and amplitudes with frequencies  $\omega_2 = \omega_{p_0} + 1\omega_r$  for  $p = 1$ ,  $\omega_2 = \omega_{p_0} + 2\omega_r$  for  $p = 2$ . The new angular momentum operator,  $|L_2, -L_2\rangle_f = |l-2\rangle_f |0\rangle_1 \dots |0\rangle_n$  represents the pump state which lost two photons from the nutation process. To find the expressions for correction,  $E_2^2$ , we must permute operators  $\check{c}_{p_1}^\dagger(t)$  from right hand to left-hand side in order to act with them on the bra-vector,  $\langle L, -L|_f \check{c}_{p_1}^\dagger \check{c}_{p_2}^\dagger = \delta_{p_2',0} \delta_{p_1',0} \sqrt{l(l-1)} \langle l-2|_0 \langle 0|_1 \dots \langle 0|_n$ . The simple observation demonstrates that this part of operator transfer in the left-hand part of correlation function,  $\langle L, -L|_f (L_n^-)^2 \check{c}_{p_2}^\dagger(t) \check{c}_{p_1}^\dagger(t)$ , is Hermit conjugate to the right-hand permutations, (37). Multiplying the Hermit conjugate expression of this amplitude,  $\langle L, -L|_f (L_n^-)^2 \check{c}_{p_2}^\dagger(t) \check{c}_{p_1}^\dagger(t)$ , with Exp. (37), we can obtain the following expression of correlation,

$$\begin{aligned} E_2^2 &= l(l-1) \{ \delta_{p_2,0} \delta_{p_1,0} \delta_{p_2',0} \delta_{p_1',0} 2^2 L_2 (2L_2 - 1) \\ &+ 8s 2L_2 (\delta_{p_2,0} \delta_{p_1-1,0} + \delta_{p_1,0} \delta_{p_2-1,0}) (\delta_{p_2',0} \delta_{p_1'-1,0} + \delta_{p_1',0} \delta_{p_2'-1,0}) \\ &+ [\sqrt{2!2s(2s-1)} (\delta_{p_1-2,0} \delta_{p_2,0} + \delta_{p_2-2,0} \delta_{p_1,0}) + 2s \delta_{p_2-1,0} \delta_{p_1-1,0}] \\ &\times [\sqrt{2!2s(2s-1)} (\delta_{p_1'-2,0} \delta_{p_2',0} + \delta_{p_2'-2,0} \delta_{p_1',0}) + 2s \delta_{p_2'-1,0} \delta_{p_1'-1,0} \}. \end{aligned}$$

Similarly, we may estimate all correlation functions in the first and second moment of intensity rate. Knowing the correlations  $E_0^1$ ,  $E_1^1$ ,  $E_2^1$  and  $E_3^1$  we may estimate the loses of excitations from the cavity, or intensity of scattering field. The quantum correlation between the converted photons may be estimated knowing the square loses (or square intensity correlations). For this we must introduce in Exp. (36) the correlations  $E_0^2$ ,  $E_1^2$ , and  $E_2^2$  in the second order moment,  $\alpha = 2$ . The correlation  $E_3^2$  is multiplied by  $K_{-1}^2 = 0$  doesn't give the contribution in the square losses of excitations from the resonator.

According to the representation (36) we obtain the following expressions for numerical simulation of the first and second moment of the cavity excitations,

$$\begin{aligned} \left\langle \frac{d}{dt} \hat{W}_e(t) \right\rangle &= G_{c1} C_{s1}(t) + G_{s1} S_{n1}(t) + G_{c2} C_{s2}(t) + G_{s2} S_{n2}(t); \\ \left\langle \left( \frac{d}{dt} \hat{W}_e(t) \right)^2 \right\rangle &= K_{c1} C_{s1}(t) + K_{s1} S_{n1}(t) + K_{c2} C_{s2}(t). \end{aligned} \quad (38)$$

Here the coefficients calculated for fist moment are:

$$\begin{aligned} G_{c1} &= 8l\Gamma_{00}; \\ G_{s1} &= 2^3 \times 3ls \{ 2\Gamma_1 + (l-1)\Gamma_0 \}; \\ G_{c2} &= 2^4 \times 3l \{ \Gamma_0 L_1 (2L_1 - 1) + 2^3 \Gamma_1 L_1 s + \Gamma_2 (2s-1)s \}; \\ G_{s2} &= 2^4 \times 3^2 l \{ L_1 (2L_1 - 1) (2L_1 - 2) \Gamma_0 + 2s L_1 (2L_1 - 1) \Gamma_1 \\ &+ 2L_1 s (2s-1) \Gamma_2 + 2s (2s-1) (s-1) \Gamma_2 \}. \end{aligned} \quad (39)$$

and for second moment we obtain,

$$\begin{aligned}
 K_{c1} &= 2^5 \times 3\Gamma_{00}^2 l(l-1); \\
 K_{s1} &= 2^4 \times 3^2 \times l(l-1) \{s[2^3\Gamma_{11}\Gamma_{00} + 3^3\Gamma_{01}\Gamma_{10}] + 2L_2\Gamma_{00}\Gamma_{00}\} \\
 K_{c2} &= 2^7 \times 3l(l-1) \{L_2(2L_2-1)\Gamma_{00}^2 + 8sL_2[\Gamma_{00}\Gamma_{11} + \Gamma_{10}\Gamma_{01}] \\
 &\quad + 4s(2s-1)[3(\Gamma_{00}\Gamma_{22} + 4\Gamma_{20}\Gamma_{02})] \\
 &\quad + 15s\sqrt{s(2s-1)}\Gamma_{01}\Gamma_{21} + 4s^2\Gamma_{11}\Gamma_{11}\}.
 \end{aligned} \tag{40}$$

The above coefficients introduced in Exps. (38) were calculated according to the correlation functions described by equations for moments (36). Their indexes are connected with cosine oscillation functions,  $C_{s1}(t)$ ;  $C_{s2}(t)$ , or sine one,  $S_{n1}(t)$ ,  $S_{n2}(t)$ . The coefficients,  $G_{c1}$  and  $K_{c1}$ , don't contain the information about the scattering steps,  $n = 2s$  in multiple scattering process. This information about cooperative multiple conversion of photons appears in the next time moments of the kinetic process, described by correlations calculated using multiple excited state,  $(\hat{\Lambda}_n^+)^l |L, -L\rangle_f$ ,  $s = 1, 2, 3$ . Such type of conversion process is contained in the definition of the coefficients,  $G_{s1}$ ;  $G_{c2}$ , and,  $K_{s1}$ ,  $K_{c2}$ . As follows from the expressions (39) and (40), these coefficients are proportional to this transfer cooperative number,  $n = 2s$ , and its moments. Here the new cooperative numbers,  $L_1 = s(l-1)$  and  $L_2 = s(l-2)$ , represent the new states of the cavity excitations from which one or two photons leave the cavity die to the scattering process in the external field. Taking into consideration that the coefficients  $\Gamma_{ij}$  depends on the interaction constants with external field,  $\chi_{kp} \sim g_{p+1}$ . According to the representation (21) we approximate the rate matrix  $\Gamma_{ij}$  from coefficients (39) and (40) by the expressions,  $\Gamma_{ij} \approx G_0 \sqrt{(j+1)(2s-j)} \sqrt{(i+1)(2s-i)}$ ,  $\Gamma_{ij} \equiv \Gamma_j$ . As follows from the numerical estimations, the cooperative correlations of scattering amplitude from different steps increases in the second order moment,  $\langle (dW_e/dt)^2 \rangle$ , with increasing of number of photons in the pump mode of the cavity. In figure a. and b. we observe that second order moment is smaller than first one,  $d\langle W_e \rangle/dt$ , when the number of photons in the pump mode is less than 15. The increasing the number of photons,  $l = 22$ , and  $l = 30$ , stimulate the coherent and cooperative process in the system C, so that relative quantum fluctuation decreases,  $\Delta_W / (d\langle W_e \rangle/dt)^2$ .

In conclusion to this section, we observe the following. The multiple scattering effect is manifested for large number of photons. To detect the superposition of multiple scattering photons in the big number of modes, it is necessary to have the large ensemble of excited atoms which may multiple scattered the pump field. As follows from the estimation of Exp. (36), for four atoms the possibility to observe the high mode correlations,  $E_3^2$ , becomes impossible due small number scattering centers (radiators) in the system. Here exist two possibilities. The first action is to, return to last section and solve exactly the Schrödinger equation for large ensemble of atoms. The second possibility is to pump the excited atoms into the resonator and obtain the multiple scattering laser. The first possibility is limited by the impossibility to solve exactly the system of operator vectors for big number of atoms or large numbers of photons in the pump field. Such a problem involves the large degree of freedoms in the proposed approach of section 2, which limits the possibilities to represent in analytic form of the solutions of Schrödinger equation. The second approach is connected to finding the system of known wave functions like,  $(\hat{\Lambda}_n^+)^k |l\rangle_0 |0\rangle_{1\dots n}$ , of multiple scattered field on Hilbert space, on which we may construct the evolution of such a quantum correlations between and possible excitations in the in the cavity bi-modes during the multiple scattered photons.

## 4. Discussions

We introduced the new field characteristics of bi-modes in the coherent state in which the multiple steps Raman lasing described by the superposition,  $\mathbf{E}^{(+)}(z, t) = \mathbf{E}_{p_0}^{(+)}(z, t) + \mathbf{E}_{as1}^{(+)}(z, t) + \mathbf{E}_{as2}^{(+)}(z, t) + \dots + \mathbf{E}_{asn}^{(+)}(z, t)$ , with frequencies of the pump, first, second, ..., and  $n$  anti-Stokes components with frequencies  $\omega_{p_0}$ ,  $\omega_{p_0} + \omega_r$ , ...,  $\omega_{p_0} + n\omega_r$ . Here  $\hbar\omega_r$  is the excitation energy of each radiator (atom, molecule, exciton). The new characteristic of bimodal excitation of the cavity field was introduced in the co-linear cavity/fiber approximation,  $\hat{\Pi}^+(t) = \hat{E}^{(+)}(z, t)\mathbf{E}^{(-)}(z, t)$ , which permit us to represent the total intensity correlation as a sum of each component:

$$\begin{aligned}
 \langle \hat{\Pi}^-(t)\hat{\Pi}^+(t) \rangle_{(n)} &= \langle \hat{\Pi}_{1n}^-(t)\hat{\Pi}_{1n}^+(t) \rangle + \langle \hat{\Pi}_{2n}^-(t)\hat{\Pi}_{2n}^+(t) \rangle \\
 &\quad + \dots + \langle \hat{\Pi}_{\alpha n}^-(t)\hat{\Pi}_{\alpha n}^+(t) \rangle + \dots
 \end{aligned} \tag{41}$$

where  $\alpha \leq n$  is over scattering steps parameter,  $\alpha \leq n$ . The first adjacent mode correlation,  $\hat{\Pi}_{1n}^+(t) \sim \sum_m^n \hat{E}_{asm}^{(+)}(t)\hat{E}_{asm-1}^{(-)}(t)$ , the second adjacent one (over two steps),  $\hat{\Pi}_{2n}^+(t) \sim \sum_m^n \hat{E}_{as1}^{(+)}(t)\hat{E}_{asn-2}^{(-)}(t)$ ; and the over  $\alpha$  - step correlation,  $\hat{\Pi}_{\alpha n}^+(t) \sim \sum_m^n \hat{E}_{asm}^{(+)}(t)\hat{E}_{asm-\alpha}^{(-)}(t)$ , represent the characteristics of the bimodal cavity field in multiple scattering process at frequencies differences,  $\omega_r$ ;  $2\omega_r$ ; ...,  $\alpha\omega_r$ . Multiplying these characteristics to Hermitconjugate components,  $\hat{\Pi}_{1m}^-(t)$ ,  $\hat{\Pi}_{2m}^-(t)$ , and  $\hat{\Pi}_{\alpha m}^-(t)$ , we observe that in Exp. (41) is described by the

moments (36) and can be observed by losses equation (11). The coherent superpositions of these components may be used as a guide field characteristic with the good phase and amplitude in the semi-classical approximation,  $\langle \hat{\Pi}_{an}^+(t) \rangle = P_{kn0} \exp[-i\phi_\alpha]$ , in the lasing process. Here, the Stokes component of the generation of bimodal field,  $\hat{E}_j^+(t)$ , can be regarded as a pump field for the next order of Raman lasing (See, for example, [37]), so that the products,  $\hat{E}_m^+(t)\hat{E}_{m-\alpha}^-(t)$ , has the same phase,  $\phi_{k\alpha} = i\alpha\omega_r t + i\alpha Kz$ , in which  $\hbar\alpha\omega_r = \hbar\omega_{j+\alpha} - \hbar\omega_j$ , and  $K = k_{j+\alpha} - k_j$  are the  $\alpha$  portions of excitation energy that the cavity field obtains when the atomic system passes from the excited to ground states in  $\alpha$  multiple Raman steps,  $\alpha = 1, 2, 3, \dots$ . Here  $K$  is the wavevector of bimodal components of induced lasing in the cavity. The detection possibilities of such cooperative phenomena after the propagation of correlated photons through different fibers may be realized using figure 5 in which instead of losses in the outside free field, the contact between the fibers and spherical cavity is proposed [39–41]. According to this approach, only the diagonal elements belonging to the same modes remain non-zero so that the field intensity is proportional to this number of steps, ‘ $n$ ’. The possibility of correlations between the anti-Stokes/Stokes components in the multiple Raman scattering have been overcome by recent advances in coherent scattering microscopy, which is based on coherent anti-Stokes/Stokes components stimulated by the pump photons [2–10].

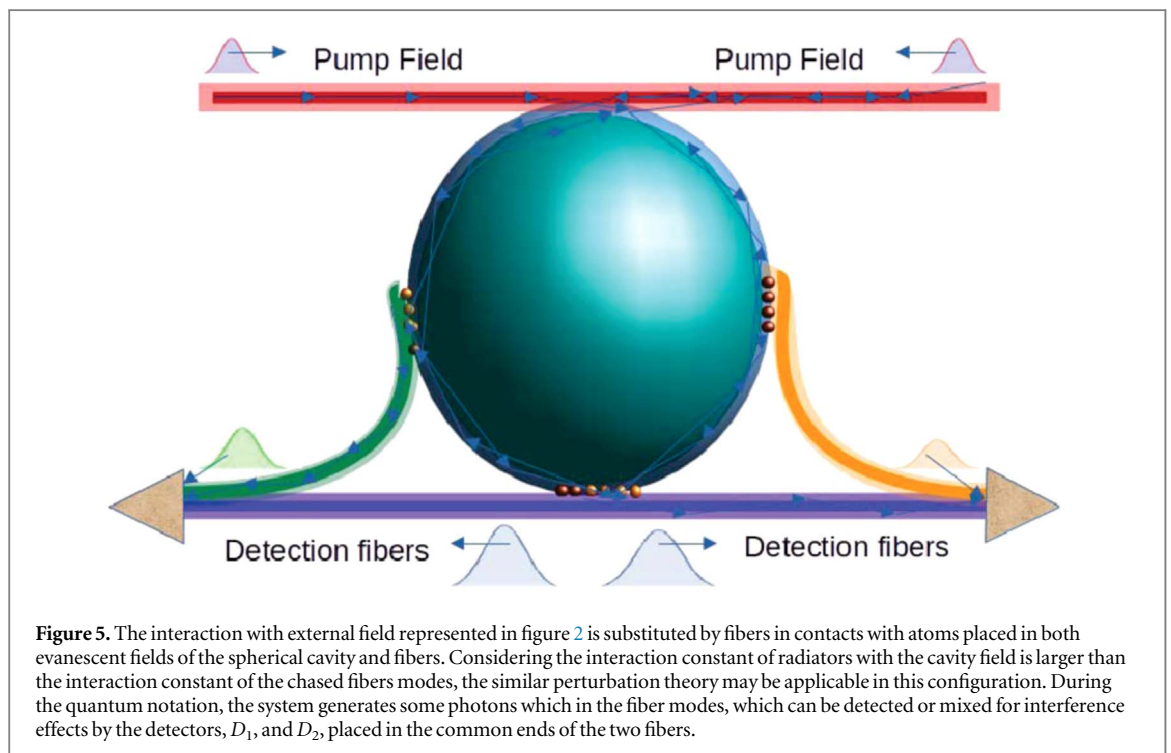
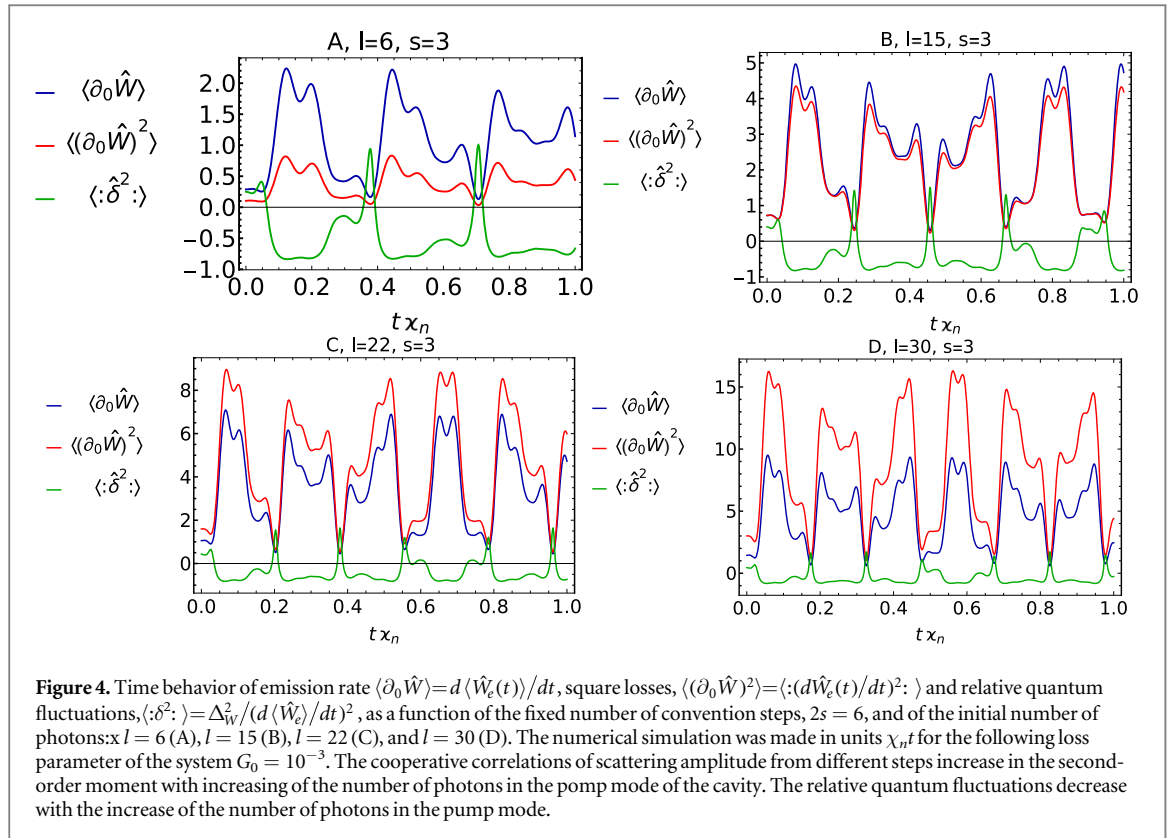
We focused our attention to quantum correlations between the components of multiple Raman scattering in which the multi-steps induce emission opens the opportunity. To understand this type of coherence, we purpose to study the inverse quantum conversion in which the atomic scattering centers (molecules, atoms, etc.) are prepared in the ground state (see figure 1B). In this situation, the modes in which are converted the pump photons in multiple scattering process belong to Stokes components,  $\omega_1 = \omega_p - \omega_r$ ,  $\omega_2 = \omega_p - 4\omega_0$ ,  $\omega_3 = \omega_p - 6\omega_0$ , etc. Such a red shift was observed in intra-cavity continuous-wave of multiple Raman scattering emissions. The application of such cooperative effects may be used in photon recycling and scattering in high-performance solar cells [42, 43]. The possible quantum nutation between two multiple scattering process represented in the figure 1 has a specific interest in quantum physics, due to the fact that portions of quanta with frequency proportional to the transition energy between the excited and ground states of radiator levels,  $\omega_r = \omega_{p+1} - \omega_p$ , periodically pass from atomic subsystem to cavity field describing quantum reversibility at short time intervals.

The moments (14) and (36) described in the sections 2 and 3 have some analogies with quantum Fisher information described in the [44–47]. In this situation, we must introduce the distribution function for the number of photons,  $\hat{\rho}(t) \sim \hat{W}_C(t)/N_f$ , and the product of two derivative functions in the second moment (14). Here the number of photons inside the resonator,  $N_f = N - N_e$  can be approximated with the total number,  $N$ , when the number of the losses photons is neglected,  $N_f > N_e$ . This procedure can be used for the construction of the quantum Fisher information form,  $F_{\mu,\nu} = \text{Tr} \{ \partial_\mu^s \hat{\rho}(\tau_\mu) \partial_\nu^s \hat{\rho}(\tau_\nu) \}$ , where  $\partial_\mu^s \hat{\rho}$  and  $\partial_\nu^s \hat{\rho}(\tau_\nu)$  represent the symmetric logarithmic derivative through the anti-commutation,  $\partial_\alpha^s \hat{\rho} = \{ \hat{L}_\alpha \hat{\rho} + \hat{\rho} \hat{L}_\alpha \} / 2$ ,  $\alpha \equiv \nu, \mu$ . Here  $\hat{L}_\mu$  and  $\hat{L}_\nu$ , operators described in [46–48]. Indeed, for large value of excitations incise the resonator,  $N_f \sim N$  we can find the correlation between the diagonal Fisher information,  $F_{\mu,\mu}$ , and our second moment (14). Considering that  $\tau_\mu \sim t\chi_\mu$ , the quantum Cram?–Rao bound is the quantum analog of the classical Cram?–Rao bound  $(\Delta\tau_\mu)^2 \geq 1/(MF_{\mu,\mu})$ . It may be estimated according to figure 4, Here  $M$  is the number of independent repetitions [49–51]. According to this expression and the numerical behavior of the second moment represented in figure 4, we observe that the quantum phase fluctuation achieved the minimal value when Fisher Information (or the second moment) achieved the maximal one. This conclusion demonstrates the possibilities to realize the coherent lasing processes of multimode scattering cooperative effect in which the quantum correlations are established not only between the adjacent mode but between non-adjacent too.

## 5. Conclusions

In this approach, we put the problem about quantum cooperative process between the photons in the multiple steps of scattering by the system of radiators. Here the conception is divided into two parts. First part corresponds to the multiple generation of photons in the  $n$ -scattered cavity modes during the quantum notation of radiators. Here we used the possibilities of transmission of energy multiple to portion  $\hbar\omega_r$  to the radiators or to the cavity field when the atomic inversion surfer the nutation. A great attention was made to the correlations not only between the adjacent mode of multiple scattering process, but to the possible quantum correlations between the photons belonging to the non-congruent steps of the multiple scattering process. We mean the correlations like,  $\langle \hat{E}_{i+s}^{(+)}(z, t) \hat{E}_i^{(-)}(z, t) \rangle$  in which the number of steps,  $i$ , and  $i + s$ , are established in the system so that correlation takes between nonadjacent modes  $s > 1$  of the multiple scattering. This quantum process opens the possibilities to transmit the information not only between the neighboring modes,  $\hat{E}_{i\pm 1}$  and  $\hat{E}_i$ , but between the next steps of multiple scattering represented in figures 2 and 5. The extension of the correlations between the non-adjacent steps in the case of the exact solutions of the quantum nutation in the Raman scattering suffers





from an impossibility to solve the Schrödinger equation for a big number of radiators or for many photons in the pumping field. This process may be solved studying the multiple scattering lasing [37]. In figure 5 we propose the possibility to detect and study the quantum correlations between the photons of non-adjacent modes. Introducing the thin fibers into evanescent zone of the cavity, we may choose the coupling of atomic subsystem with cavity larger than coupling with the modes of thin fibers. These possible schemes of riding the information from micro-cavities using thin fibers are in the center of interests in many modern experiments [39, 41–52].



## Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).  
<https://doi.org/yes>.

## Exact solutions for: (a) small number of atoms and arbitrary bimodal excitations; (a) small number of bimodal excitations and arbitrary number of atoms

In this appendix, we are focused on the possibilities to construct the closed system of the linear operator equations like (19) the determinate of which contains commutative terms so that we can use the method of the solution described in the literature.

Let's start with a small number of atoms taking the part into multiple scattering process. According to the definition of multiple scattering operators,  $\hat{\Lambda}_n^-$  and  $\hat{\Lambda}_n^+$ , we observe that they satisfy the commutation relations:

$$[\hat{\Lambda}_n^+, \hat{\Lambda}_n^-] = \sum_{p=0}^{n-1} g_{p+1}^2 \{\hat{c}_{p+1}\hat{c}_{p+1}^\dagger - \hat{c}_p\hat{c}_p^\dagger\} / \chi_n^2 = 2\Delta\hat{\Lambda}_n; \quad (\text{A } 1)$$

$$[\Delta\hat{\Lambda}_n, \hat{\Lambda}_n^+] = \sum_{p=0}^{n-1} \{g_{j+1}^2 + \theta(j-1+0.5)g_j^2/2 - g_{j+2}^2\theta(n-2-j+0.5)/2\} g_{j+1}\hat{c}_j\hat{c}_{j+1}^\dagger / \chi_n^3, \quad (\text{A } 2)$$

where the diagonal elements,  $\Delta\hat{\Lambda}_n = \sum_{p=0}^{n-1} g_{p+1}^2 \{\hat{c}_{p+1}\hat{c}_{p+1}^\dagger - \hat{c}_p\hat{c}_p^\dagger\} / (2\chi_n^2)$ , commutes with free parts of Hamiltonian (2), the normalized coefficient,  $\chi_n$ , is choosing according to the possibilities of the reducing of the generation process in the multiple scattering to known symmetry;  $\theta(x)$  is the Heaviside step function with definition:  $\theta(x) = 1$ , for  $x > 0$ ,  $\theta(x) = 1/2$ , for  $x = 0$ , and  $\theta(x) = 0$ , for  $x < 0$ . Let us return to the solutions of the system of equations (19). For two atoms,  $j = 1$ , we have three equations of atomic operator vectors,  $\hat{X}_{-1}(t)$ ,  $\hat{X}_0(t)$  and  $\hat{X}_1(t)$ , in the system (19). The equation for  $\hat{X}_0(t)$  can be reduced to the cubic differential equation,

$$\frac{d^3}{dt^3}\hat{X}_0(t) = -\frac{d}{dt}\hat{X}_0(t) \{2\chi_n^2[\hat{\Lambda}_n^+\hat{\Lambda}_n^- + \hat{\Lambda}_n^-\hat{\Lambda}_n^+] + \delta^2\} + 2i\delta\chi_n^2\hat{X}_0(t)[\hat{\Lambda}_n^+\hat{\Lambda}_n^- - \hat{\Lambda}_n^-\hat{\Lambda}_n^+].$$

The solution of this equation can be found under the form,  $\hat{X}_0(t) = \hat{C} \exp[i\hat{\Omega}t]$ , from which follows the characteristic equation,  $\hat{\Omega}^3 + \hat{\Omega}\hat{p} + \hat{q} = 0$ , where the coefficients are  $\hat{p} = -\{2\chi_n^2[\hat{\Lambda}_n^+\hat{\Lambda}_n^- + \hat{\Lambda}_n^-\hat{\Lambda}_n^+] + \delta^2\}$  and  $\hat{q} = 2\delta\chi_n^2[\hat{\Lambda}_n^+\hat{\Lambda}_n^- - \hat{\Lambda}_n^-\hat{\Lambda}_n^+]$ . The discriminant of this cubic equation is  $\hat{\Delta} = -4\hat{p} - 27\hat{q}$ . If the mean value of discriminant is positive,  $\Delta > 0$ , the cubic equation has three distinct real roots, and according to the definition of quantum Rabi frequency,  $\hat{\Omega}$ , the quantum process becomes oscillatory. For the negative values of the discriminant,  $\Delta < 0$ , the cubic equation has one real root and two complex conjugate roots, which correspond to attenuation or increasing of the amplitude of oscillatory process as function of value and the sign of the detuning from resonance  $\delta$ . According to the canonical form, the roots of characteristic equation are  $\hat{\Omega}_1 = \hat{A} + \hat{B}$ ,  $\hat{\Omega}_{2,3} = (\hat{A} + \hat{B})/2 \pm i\sqrt{3}(\hat{A} - \hat{B})/2$ , where  $\hat{A} = \sqrt[3]{-\hat{q}/2 + \sqrt{-\hat{\Delta}}}$ , and  $\hat{B} = \sqrt[3]{-\hat{q}/2 - \sqrt{-\hat{\Delta}}}$ . As follows from this equation and (19) for zero value of detuning,  $\delta = 0$ , the solution of the system of equation (19) is:

$$\begin{aligned} \hat{X}_1(t) &= -i\chi_n\sqrt{2} \{C_1 \sin[\hat{\Omega}_n t] - \hat{C}_2 [\cos[\hat{\Omega}_n t] - 1]\} \hat{\Omega}_n^{-1} \hat{\Lambda}_n^+ + |1, 1\rangle, \\ \hat{X}_0(t) &= \hat{C}_1 \cos[\hat{\Omega}_n t] + \hat{C}_2 \sin[\hat{\Omega}_n t], \\ \hat{X}_{-1}(t) &= -i\chi_n\sqrt{2} \{C_1 \sin[\hat{\Omega}_n t] - \hat{C}_2 (\cos[\hat{\Omega}_n t] - 1)\} \hat{\Omega}_n^{-1} \hat{\Lambda}_n^- + |1, -1\rangle \end{aligned} \quad (\text{A } 3)$$

where  $\hat{C}_1 = |1, 0\rangle_r$ ,  $\hat{C}_2 = -i\chi_n\sqrt{2} \{|1, 1\rangle_r \hat{\Lambda}_n^- + |1, -1\rangle_r \hat{\Lambda}_n^+\} \hat{\Omega}_n^{-1}$  and  $\hat{\Omega}_n = \chi_n\sqrt{2(\hat{\Lambda}_n^+\hat{\Lambda}_n^- + \hat{\Lambda}_n^-\hat{\Lambda}_n^+)}$ .

It is not so difficult to observe, that the substitution of the vectors in the solution (18) is reduced to Jaynes–Cummings model with nonlinear interaction of an atom with the field. We are interested in the use of symmetry in the bi-boson interaction of atoms with scattering field described by interaction Hamiltonian (3) in order to simplify the solutions of the system of equations (19). We observe that this system of equations has a non-commutative element in the main determinant, so that we meet not so simple procedure to find the eigenvectors of such a system of operator equations.

If the multiple scattering transitions in the atomic subsystem are defined without intrinsic symmetry between the field operators,  $\hat{\Lambda}_n^+$  and  $\hat{\Lambda}_n^-$ , described by the commutation relations (A1) and (A2), we don't have the possibility to obtain the solutions for close system of equations for vector operators,  $\{\hat{R}_m(t) = \hat{X}_m(\hat{\Lambda}_n^+)^{j-m}\}$  for the large number of atoms. Due to the fact that in the second term of each equation of the system, (19),  $\hat{X}_{m+1}(t)\hat{\Lambda}_n^-(\hat{\Lambda}_n^+)^{j-m}$  must be commuted according to algebraic rule. In this situation, the permutation of the operator ( $\hat{\Lambda}_n^-$ ), over  $(\hat{\Lambda}_n^+)^{j-m}$ , leaves the group symmetry in the general representation described by commutation relations (A1) and (A2). Introducing the restriction to the scattering amplitudes,  $g_b$ , of multi-step processes we can reduce this permutation to  $su(2)$  symmetry, (21) or to  $su(1, 1)$  commutation requirement (26). In this situation, the system of operator equations (19) becomes solvable for a large number of atoms using the determinant representation described above,

$$\begin{aligned} \frac{d}{dt}\hat{R}_m(t) &= -i\delta m\hat{R}_m(t) - i\chi_n\sqrt{(j+m+1)(j-m)}\hat{R}_{m+1}(t)\Delta_{j-m}^{su} \\ &\quad - i\chi_n\sqrt{(j+m)(j-m+1)}\hat{R}_{m-1}(t), \\ m &= -j, -j+1, \dots, j-1, j, \end{aligned} \quad (\text{A } 4)$$

The right-hand side of this system of equations can be represented through a vector form transformed by operator-matrix,  $\hat{D}^{(2j+1)}$

$$\frac{d}{dt}\{\hat{R}_j(t), \hat{R}_{j-1}(t), \dots, \hat{R}_{-j}(t)\} = -i\{\hat{R}_j(t), \hat{R}_{j-1}(t), \dots, \hat{R}_{-j}(t)\}\hat{D}^{(2j+1)}, \quad (\text{A } 5)$$

the definition of which depends on the applied algebra to the field operators. Here, the operator-matrix  $\hat{D}^{(2j+1)}$  has the rang  $2j+1$  and can be represented by the expression

$$\hat{D}^{(2j+1)} = \begin{pmatrix} -\delta j & \chi_n\sqrt{2j}\Delta_1^{su} & 0 & \dots & 0 & \dots \\ \chi_n\sqrt{2j} & \delta\{-j+1\} & \chi_n\sqrt{2(2j-1)}\Delta_1^{su} & \dots & 0 & \dots \\ 0 & \chi_n\sqrt{2(2j-1)} & \delta\{-j+2\} & \dots & 0 & \dots \\ \dots & \dots & \dots & \dots & \dots & \dots \\ 0 & 0 & 0 & \dots & \chi_n\sqrt{(j+m+1)(j-m)}\Delta_{j-m}^{su} & \dots \\ 0 & 0 & 0 & \dots & -\delta(L-p) & \dots \\ 0 & 0 & 0 & \dots & \chi_n\sqrt{(j+m)(j-m+1)} & \dots \\ \dots & \dots & \dots & \dots & \dots & \dots \end{pmatrix}$$

Here as function of symmetry the permutation,  $\hat{\Lambda}_n^-(\hat{\Lambda}_n^+)^{j-m} = (\hat{\Lambda}_n^+)^{j-m-1}\Delta_{j-m}^{su}$ , was estimated for  $su(2)$ , and  $su(1, 1)$  symmetries,

$$\Delta_p^{su} = \begin{cases} \hat{\Lambda}_n^-\hat{\Lambda}_n^+ - p(p-1) - 2(p-1)\hat{\Lambda}_{nz}, & \text{for } su(2); \\ \hat{\Lambda}_n^+\hat{\Lambda}_n^- + p(p-1) - 2p\hat{\Lambda}_z & \text{for, } n \rightarrow \infty \quad su(1, 1). \end{cases} \quad (\text{A } 6)$$

According to this representation (A6), the system of equation (A4) can be represented in vector-form (A5) for both symmetries.

2. The similar approach is used relative to the system of equation (24). In order to obtain the determinant with commutative elements, we propose the following substitutions in the system of equations (21),  $\hat{Y}_p(t) = \hat{F}_p(t)(\hat{D}^-)^p$ . In the real system of coordinate, this means that we pass to new variables,  $\{\hat{Y}_p(t)\}$ , in which the excitation with conversion energy,  $\hbar\omega = \hbar(\omega_{j+1} - \omega_j)$ , is lowered by atomic operator,  $\hat{D}^-$ . Observing the identity between the atomic operators,  $\hat{F}_{p-1}(t)\hat{D}^+(\hat{D}^-)^p = \hat{Y}_{p-1}(t)\hat{\Delta}_p$ , where  $\hat{\Delta}_p = \hat{D}^+\hat{D}^- + 2(p-1)\hat{D}_z - p(p-1)$ , we may easily pass from the system vectors,  $\hat{F}_p(t)$ , to the new one,  $\{\hat{Y}_p(t)\}$ , representing the system of equation (24) through the new operator system,

$$\begin{aligned} & \frac{d}{dt}(\hat{Y}_0(t) \ \hat{Y}_1(t) \ \dots \ \hat{Y}_p(t) \ \dots \ \hat{Y}_{2L}(t)) \\ & = -i(\hat{Y}_0(t) \ \hat{Y}_1(t) \ \dots \ \hat{Y}_p(t) \ \dots \ \hat{Y}_{2L}(t))\hat{D}^{(2L+1)}, \end{aligned} \tag{A 7}$$

in which the elements of the  $2L + 1$  - rang matrix,  $\hat{D}^{(2L+1)}$ , become commutative,

$$\hat{D}^{(2L+1)} = \begin{pmatrix} -\delta L & \chi_n \sqrt{2L} \hat{\Delta}_1 & 0 & \dots & 0 & \dots \\ \chi_n \sqrt{2L} & -\delta(L-1) & \chi_n \sqrt{2(2L-1)} \hat{\Delta}_2 & \dots & 0 & \dots \\ 0 & \chi_n \sqrt{2(2L-1)} & -\delta(L-2) & \dots & 0 & \dots \\ \dots & \dots & \dots & \dots & \dots & \dots \\ 0 & 0 & 0 & \dots & \chi_n \sqrt{p(2L-p+1)} \hat{\Delta}_p & \dots \\ 0 & 0 & 0 & \dots & -\delta(L-p) & \dots \\ 0 & 0 & 0 & \dots & \chi_n \sqrt{(p+1)(2L-p)} & \dots \\ \dots & \dots & \dots & \dots & \dots & \dots \end{pmatrix}.$$

In this situation, we may easily use the classical algorithm for the possible solutions of the linear system of equations (A7). The procedure of simplification of multiple scattering process help us to observe some quantum peculiarities in the behaviors of emitted photons taking into consideration relative not small number of photons or atoms, described by the system of equation like (A7). The solution of the system of equation (A7) can be represented by vectors in the form  $\{\hat{Y}_p(t) = \hat{Y}_p(0) \exp[i\hat{\Omega}t]\}$ ,  $p = 0, 1, 2, \dots, n$ . The requirement of the matrix diagonalization,  $\det \hat{M}^{(2L+1)} = 0$ , where  $\hat{M}^{(2L+1)} = \hat{D}^{(2L+1)} + \hat{I}\hat{\Omega}$ , drastically simplifies the solution of the system of equation, (A7) (here  $\hat{I}$  is the identity matrix of size  $2L + 1$ ). From this procedure of diagonalization, we may formally find the ‘eigenvalues’,  $\hat{\Omega}_p$ .

Below we take some example how to use this formalism in the solution of the system of equations (A7) for detuning,  $\delta = 0$ . We observe that two-step Raman conversion appear for the cooperative number,  $L = 1$ , from which follows two possibilities: first corresponds to traditional single step Raman conversion,  $s = n/2 = 1/2$ , and two photons in the pump mode,  $l = 2$ ; and second situation two-step multiple Raman conversion  $s = 1$  of a single photon in pump mode,  $l = 1$ . The second one contains the possibility to convert initial photon state  $|1\rangle_0|0\rangle_1|0\rangle_2$  into anti-Stokes,  $|0\rangle_0|1\rangle_1|0\rangle_2$ , and after that use the anti-Stokes photon for next step of conversion into state,  $|0\rangle_0|0\rangle_1|1\rangle_2$ . In this situation the procedure of diagonalization of the matrix,  $\hat{D}^{(3)}$ , gives us the following equation for eigenvalues,  $\hat{\Omega}^3 - 2\chi_n(\hat{\Delta}_2 + \hat{\Delta}_1)\hat{\Omega} = 0$ , the solutions of which are reduced to:  $\hat{\Omega}_0 = 0, \hat{\Omega}_{1,2} = \pm g\sqrt{2(\hat{\Delta}_2 + \hat{\Delta}_1)}$ .

Let us take, the cooperative number equal to  $L = ln/2 = 3/2$ . We also observe two possibilities in the realization of this total cooperative number: (a) the number of photons in pump field is equal to three,  $l = 3$ , in the single step Raman scattering,  $n = 1$  ( $s = 1/2$ ); (b) we may have one photon in pump field and,  $l = 1$ , and three scattering steps may be realized,  $n = 3$ , which corresponds to  $s = 3/2$ . According to this from the equation for the matrix diagonalization,  $\det[\hat{D}^{(4)} + \hat{I}\hat{\Omega}] = 0$ , we obtain the following algebraic equation for eigenvalues,

$$\hat{\Theta}^4 - \hat{\Theta}^2(3\hat{\Delta}_3 + 4\hat{\Delta}_2 + 3\hat{\Delta}_1) + 9\hat{\Delta}_1\hat{\Delta}_3 = 0,$$

from which follows four solutions for Rabi frequencies. Below we represent them in pairs,

$$(\hat{\Theta}^2)_{1,2} = \frac{3\hat{\Delta}_3 + 4\hat{\Delta}_2 + 3\hat{\Delta}_1}{2} \pm \frac{1}{2}\sqrt{(3\hat{\Delta}_3 + 4\hat{\Delta}_2 + 3\hat{\Delta}_1)^2 - 36\hat{\Delta}_1\hat{\Delta}_3}. \tag{A 8}$$

Here  $\hat{\Theta}_i = \hat{\Omega}_i/\chi_n$ ,  $i = 1, 2, 3, 4$ , is the Rabi frequency in the relative units in which two of them is expressed through positive sign in the expression (A8),  $\hat{\Theta}_{1,3} = \pm\sqrt{(\hat{\Theta}^2)_1}$ , and second pair is connected with negative sign in the same relation (A8),  $\hat{\Theta}_{2,4} = \pm\sqrt{(\hat{\Theta}^2)_2}$ .

We observe that for fixed common cooperative number,  $L$ , when we take into consideration a big number of steps in multiple Raman scattering, according to the definition,  $L=s$ , the number of photons in the pump field must be small. For example for,  $L = 2$ , we have a possibility to chose two photons in the pump field,  $l = 2$  and two steps,  $s = n/2 = 1$ , but of course this case for a single photon in the pump field we may have the four steps,  $s = n/2 = 2$ , with the multiple conversions of the photon between the fife states of cavity field. Here we consider the four non-zero interaction constants,  $g_1, g_2, g_3$  and  $g_4$ . For five operator—vectors we have fife solution of characteristic equation,  $\text{Det} \{ \hat{M}^{(5)} \} = 0$ , described by the algebraic equation,

$$\hat{\Theta}[\hat{\Theta}^4 - 2\hat{\Theta}^2(2\hat{\Delta}_1 + 3\hat{\Delta}_2 + 3\hat{\Delta}_3 + 2\hat{\Delta}_4) + 8(3\hat{\Delta}_2\hat{\Delta}_4 + 3\hat{\Delta}_1\hat{\Delta}_3 + 2\hat{\Delta}_1\hat{\Delta}_4)] = 0.$$

The relative quantum Rabi frequencies are:  $\hat{\Theta}_0 = 0$ , and four non zero solutions follows from the expression,

$$(\hat{\Theta}^2)_{1,2} = 2\hat{\Delta}_1 + 3\hat{\Delta}_2 + 3\hat{\Delta}_3 + 2\hat{\Delta}_4 \pm \sqrt{(2\hat{\Delta}_1 + 3\hat{\Delta}_2 + 3\hat{\Delta}_3 + 2\hat{\Delta}_4)^2 - 8(2\hat{\Delta}_4\hat{\Delta}_1 + 3\hat{\Delta}_4\hat{\Delta}_2 + 3\hat{\Delta}_3\hat{\Delta}_1)}. \quad (\text{A } 9)$$

Here the next Rabi frequencies,  $\hat{\Omega}_i = \hat{\Theta}_i$ ,  $i = 1, 2, 3, 4$ , are obtained from the expression (A9) in the same way as in the case for cooperative number  $L = 3/2$ .

Now we construct eigenvectors for each case described above. According to the characteristic equations, for  $L = 1$  we obtain the following solution of the system of equation (A7),

$$\begin{aligned} \hat{Y}_0(t) &= -i\chi_2\sqrt{2}|1, 0\rangle_f \hat{D}^- \hat{\Omega}_1^{-1} \sin(\hat{\Omega}_1 t) \\ &\quad + 2\chi_2^2\{|1, -1\rangle_f \hat{D}^+ \hat{D}^- + |1, 1\rangle_f (\hat{D}^-)^2\} \hat{\Omega}_1^{-2} [\cos(\hat{\Omega}_1 t) - 1] + |1, -1\rangle_f; \\ \hat{Y}_1(t) &= -i\sqrt{2}\chi_2\{|1, -1\rangle_f \hat{D}^+ \hat{D}^- + |1, 1\rangle_f (\hat{D}^-)^2\} \hat{\Omega}_1^{-1} \sin(\hat{\Omega}_1 t) \\ &\quad + |1, 0\rangle_f \hat{D}^- \cos(\hat{\Omega}_1 t); \\ \hat{Y}_2(t) &= -i\chi_2\sqrt{2}|1, 0\rangle_f \hat{D}^- \hat{\Omega}_1^{-1} \sin(\hat{\Omega}_1 t) (\hat{D}^+ \hat{D}^- + 2\hat{D}_z - 2) \\ &\quad + 2\chi_2^2\{|1, -1\rangle_f \hat{D}^+ \hat{D}^- + |1, 1\rangle_f (\hat{D}^-)^2\} \hat{\Omega}_1^{-2} [\cos(\hat{\Omega}_1 t) - 1] \\ &\quad + |1, 1\rangle_f (\hat{D}^-)^2. \end{aligned} \quad (\text{A } 10)$$

Here,  $\hat{\Omega}_1 = \chi_2\sqrt{\hat{D}^+ \hat{D}^- + \hat{D}_z - 1}$  is the quantum Rabi frequency for cooperative number,  $L=1$ . For one photon we have three migration stats:  $|1, 1\rangle_f = |1\rangle_0 |0\rangle_1 |0\rangle_2$ ;  $|1, 0\rangle_f = |0\rangle_0 |1\rangle_1 |0\rangle_2$ , and  $|1, -1\rangle_f = |0\rangle_0 |0\rangle_1 |1\rangle_2$ . We may easily pass from the system of vectors  $\hat{Y}_0(t)$ ,  $\hat{Y}_1(t)$ ,  $\hat{Y}_2(t)$  to old system operators  $\hat{F}_0(t)$ ,  $\hat{F}_1(t)$ ,  $\hat{F}_2(t)$  of the system (24). For this we must make the permutation of operator  $\hat{D}^-$  in the end part of each term of the solutions (A10) using the identity,  $\hat{D}^- \hat{\Omega}_1^2 = \hat{\Omega}_0^2 \hat{D}^-$ , where operator,  $\hat{\Omega}_0 = \chi_2\sqrt{\hat{D}^+ \hat{D}^- + \hat{D}^- \hat{D}^+}$ , plays the role form of Rabi frequency as in the solutions (A3) of the system of equations (19) for two atom subsystems in interaction with multiple scattering field. It is evidently that in the two-step Raman conversion have the solutions similar to Exps. (A3), but with substitutions,  $\hat{\Lambda}_n^+ \rightarrow \hat{D}^-$  and  $\hat{\Lambda}_n^- \rightarrow \hat{D}^+$ . This demonstrates that the proposed method of ‘eigenvalues’ and ‘eigenvectors’ in the operator form, is applicable in quantum physics. If this method gives the plausible solutions, we continue to use the method of construction of eigenvectors for cooperative numbers,  $L = 3/2$ , and  $L = 2$ .

For  $L = 3/2$  we present only one operator vector,  $\hat{Y}_0(t)$ , considering that using the initial conditions follows from the system of equations (A9) we may easily construct the other vectors,

$$\hat{Y}_0(t) = \hat{A}_1 \cos(\hat{\Omega}_1 t) + \hat{A}_2 \cos(\hat{\Omega}_2 t) + \hat{B}_1 \sin(\hat{\Omega}_1 t) + \hat{B}_2 \sin(\hat{\Omega}_2 t),$$

where the coefficients of cosine functions are expressed through the atomic operators,

$$\begin{aligned} \hat{A}_1 &= \{|3/2, -3/2\rangle_f (\hat{\Omega}_2^2 - 3\chi_3^3 \hat{\Delta}_1) - 2\sqrt{3}\chi_3^2 |3/2, 1/2\rangle_f (\hat{D}^-)^2\} / (\hat{\Omega}_2^2 - \hat{\Omega}_1^2); \\ \hat{A}_2 &= \{|3/2, -3/2\rangle_f (\hat{\Omega}_1^2 - 3\chi_3^3 \hat{\Delta}_1) (\hat{\Omega}_1^2 - 3\chi_3^3 \hat{\Delta}_1) \\ &\quad - 2\sqrt{3}\chi_3^2 |3/2, 1/2\rangle_f (\hat{D}^-)^2\} / (\hat{\Omega}_1^2 - \hat{\Omega}_2^2); \end{aligned}$$

and coefficients of sine functions are represented through similar operators,

$$\begin{aligned} \hat{B}_1 &= i\chi_3\{\sqrt{3}|3/2, -1/2\rangle_f \hat{D}^- [\hat{\Omega}_2^2 - \chi_3^2(3 + 4\hat{\Delta}_2)] \\ &\quad + 6\chi_3^2 |3/2, 3/2\rangle_f (\hat{D}^-)^3\} / [\hat{\Omega}_1(\hat{\Omega}_2^2 - \hat{\Omega}_1^2)]; \\ \hat{B}_2 &= i\chi_3\{\sqrt{3}|3/2, -1/2\rangle_f \hat{D}^- [\hat{\Omega}_1^2 - \chi_3^2(3 + 4\hat{\Delta}_2)] \\ &\quad + 6\chi_3^2 |3/2, 3/2\rangle_f (\hat{D}^-)^3\} / [\hat{\Omega}_2(\hat{\Omega}_1^2 - \hat{\Omega}_2^2)]. \end{aligned}$$

We observe only two possibilities of distribution between two cooperative numbers,  $l$ , and  $s$ : for  $l = 3, s = 1/2$  we have the states,  $|3/2, 3/2\rangle_f = |3\rangle_0, |0\rangle_1$ ;  $|3/2, 1/2\rangle_f = |2\rangle_0, |1\rangle_1$ ;  $|3/2, -1/2\rangle_f = |1\rangle_0, |2\rangle_1$  and  $|3/2, -3/2\rangle_f = |0\rangle_0, |3\rangle_1$  or for  $l = 1, s = 3/2$ , we observe the migration of photon between the four states,  $|3/2, 3/2\rangle_f = |1\rangle_0, |0\rangle_1 |0\rangle_2 |0\rangle_3$ ;  $|3/2, 1/2\rangle_f = |0\rangle_0, |1\rangle_1 |0\rangle_2 |0\rangle_3$ ;  $|3/2, -1/2\rangle_f = |0\rangle_0, |0\rangle_1 |1\rangle_2 |0\rangle_3$ ;  $|3/2, -3/2\rangle_f = |0\rangle_0, |0\rangle_1 |0\rangle_2 |1\rangle_3$ .

For  $L = 2$  we obtain five steps and five operator - ‘eigenvalues’ which give us the possibility to represent the vector in the form,

$$\hat{Y}_0(t) = \hat{A}_0 + \hat{A}_1 \cos(\hat{\Omega}_1 t) + \hat{A}_2 \cos(\hat{\Omega}_2 t) + \hat{B}_1 \sin(\hat{\Omega}_1 t) + \hat{B}_2 \sin(\hat{\Omega}_2 t), \quad (\text{A } 11)$$

where the free coefficient is

$$\begin{aligned} \hat{A}_0 = & |2, -2\rangle_f - 2^3 \chi_4^4 \{ |2, -2\rangle_f \hat{\Delta}_1 (3\hat{\Delta}_3 + 2\hat{\Delta}_4) \\ & + \sqrt{6} |2, 0\rangle_f (\hat{D}^-)^2 \hat{\Delta}_4 - 3 |2, 2\rangle_f (\hat{D}^-)^4 \} \hat{\Omega}_1^{-2} \hat{\Omega}_2^{-2}, \end{aligned} \quad (\text{A } 12)$$

and cosine coefficients may be expressed through  $\hat{A}_0$ ,

$$\begin{aligned} \hat{A}_1 = & \{ |2, -2\rangle_f (4\chi_4^2 \hat{D}^+ \hat{D}^- - \hat{\Omega}_2^2) + 2\sqrt{6} \chi_4^2 |2, 0\rangle_f (\hat{D}^-)^2 + \hat{A}_0 \hat{\Omega}_2^2 \} / (\hat{\Omega}_1^2 - \hat{\Omega}_2^2); \\ \hat{A}_2 = & \{ |2, -2\rangle_f (4\chi_4^2 \hat{D}^+ \hat{D}^- - \hat{\Omega}_1^2) + 2\sqrt{6} \chi_4^2 |2, 0\rangle_f (\hat{D}^-)^2 + \hat{A}_0 \hat{\Omega}_1^2 \} / (\hat{\Omega}_2^2 - \hat{\Omega}_1^2). \end{aligned} \quad (\text{A } 13)$$

The sine coefficients are

$$\begin{aligned} \hat{B}_1 = & 2i\chi_4 \{ |2, -1\rangle_f \hat{D}^- [\hat{\Omega}_2^2 - 2\chi_n^2 (2\hat{\Delta}_1 + 3\hat{\Delta}_2)] \\ & - 6\chi_4^2 |2, 1\rangle_f (\hat{D}^-)^3 \} / [\hat{\Omega}_1 (\hat{\Omega}_1^2 - \hat{\Omega}_2^2)]; \\ \hat{B}_2 = & 2i\chi_4 \{ |2, -1\rangle_f \hat{D}^- [\hat{\Omega}_1^2 - 2\chi_n^2 (2\hat{\Delta}_1 + 3\hat{\Delta}_2)] \\ & - 6\chi_4^2 |2, 1\rangle_f (\hat{D}^-)^3 \} / [\hat{\Omega}_2 (\hat{\Omega}_2^2 - \hat{\Omega}_1^2)]. \end{aligned} \quad (\text{A } 14)$$

The possibilities of the distribution between two cooperative numbers for same  $L$  increases. For  $L = 2$  we have three possibilities:  $l = 4, s = 1/2$  with cooperative numbers,  $|2, 2\rangle_f = |4\rangle_0, |0\rangle_1; |3\rangle_0, |1\rangle_1; \dots |0\rangle_0, |4\rangle_1$   $l = 2, s = 1$ , with two-step vectors conversion,  $|2, 2\rangle_f = |2\rangle_0, |0\rangle_1 |0\rangle_2; |2, 1\rangle_f = |1\rangle_0, |1\rangle_1 |0\rangle_2; |2, 0\rangle_f \sim |1\rangle_0, |0\rangle_1 |1\rangle_2$  or  $|0\rangle_0, |2\rangle_1 |0\rangle_2; |2, -1\rangle_f = |0\rangle_0, |1\rangle_1 |1\rangle_2; |2, -2\rangle_f = |0\rangle_0, |0\rangle_1 |2\rangle_2$ ; and  $l = 1, s = 2$ , with four steps conversion of one photon,  $|2, 2\rangle_f = |1\rangle_0, |0\rangle_1 |0\rangle_2 |0\rangle_3, |0\rangle_4; |2, 1\rangle_f = |0\rangle_0, |1\rangle_1 |0\rangle_2 |0\rangle_3, |0\rangle_4, \dots; |2, -2\rangle_f = |0\rangle_0, |0\rangle_1 |0\rangle_2 |0\rangle_3, |1\rangle_4$ . We observe that collective state,  $|2, 0\rangle$ , into two-step conversion must be the mixture of Dicke and multiple conversion states.

We may reduce the system of equation (27) for bimodal field of multiple scattering belonging to  $su(1, 1)$  symmetry (26) to a new system with commutative coefficients. In order to obtain the commutative elements of principal matrix of this system of equation, let us introduced the new vectors,  $\{ \hat{\Upsilon}_p(t) = \hat{\Phi}_p(t) (\hat{D}^-)^p \}$ , which help us to obtain a new system of equations,

$$d(\hat{\Upsilon}_0(t), \hat{\Upsilon}_1(t), \dots, \hat{\Upsilon}_p(t), \dots) / dt = -i(\hat{\Upsilon}_0(t), \hat{\Upsilon}_1(t), \dots, \hat{\Upsilon}_p(t), \dots) \hat{D}^{(\infty)}. \quad (\text{A } 15)$$

Here the principal matrix contains commutative elements,

$$\hat{D}^{(\infty)} = \begin{pmatrix} -\delta\kappa & \chi\sqrt{2\kappa} \hat{\Delta}_1 & 0 & \dots & 0 & \dots \\ \chi_n \sqrt{2\kappa} & -\delta\{\kappa + 1\} & \chi\sqrt{2((2\kappa + 1))} \hat{\Delta}_2 \dots & & 0 & \dots \\ 0 & \chi_n \sqrt{2(2\kappa + 1)} & -\delta(\kappa + 2) & \dots & 0 & \dots \\ \dots & \dots & \dots & \dots & \dots & \dots \\ 0 & 0 & 0 & \dots & \chi\sqrt{p((2\kappa + p - 1))} \hat{\Delta}_p & \dots \\ 0 & 0 & 0 & \dots & -\delta\{\kappa + p\} & \dots \\ 0 & 0 & 0 & \dots & \chi\sqrt{(p + 1)(2\kappa + p)} & \dots \\ \dots & \dots & \dots & \dots & \dots & \dots \end{pmatrix}.$$

We observe that knowing the transition symmetry, we may simplify the solution of Schrödinger equation representing it by one of three systems of equations (19), (24) and (27) described above.

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