Quantum Theory for Generation of Nonclassical Photon Pairs by a Medium with Coherent Atomic Memory

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We present a fully quantum mechanical treatment of recent experiments on creation of collective quantum memory and generation of non-classically correlated photon pairs from an atomic ensemble via the protocol of Duan et al. [Nature 414, 413 (2001)]. The temporal evolution of photon numbers, photon statistics and cross-correlation between the Stokes and anti-Stokes fields is found by solving the equation of motion for atomic spin-wave excitations. We consider a low-finesse cavity model with collectively enhanced signal-to-noise ratio, which remains still considerably large in the free-space limit. Our results describe analytically the dependence of quantum correlations on spin decoherence time and time-delay between the write and read lasers and reproduce the observed data very well including the generated pulse shapes, strong violation of Cauchy-Schwarz inequality and conditional generation of anti-Stokes single-photon pulse. The theory we developed may serve as a basic approach for quantum description of storage and retrieval of quantum information, especially when the statistical properties of non-classical pulses are studied.

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I. INTRODUCTION

For transfer of quantum states between different nodes of quantum networks, the obvious choice is to employ photons as the fast and robust carriers of quantum information [1]. However, future development of quantum communication intimately depends on successful attempts to reduce strongly the photon losses at large distances that limit the range of application of this technique. One possible way to cover large distances is using quantum repeaters [2], which combine the teleportation of entangled states as a channel of quantum information transfer (entanglement swapping) [3,4] with local atomic memories for quantum information. Among the various schemes for physical implementation of quantum repeaters and, hence, for realization of scalable long-distance communication [5–7], one of the most promising approach is the protocol of Duan, Lukin, Cirac and Zoller (DLCZ) [7]. This proposal is a probabilistic scheme that relies upon entanglement between distant atomic ensembles, which is created via successful detection of single photons emitted by initially indistinguishable sources.

In this scheme, by utilizing simple linear optical operations, a quantum repeater protocol with built-in atomic memory and entanglement purification is implemented, which is robust against the realistic imperfections, such as spontaneous loss and coupling inefficiency, that enables one to overcome photon attenuation and to communicate quantum states over arbitrary long distances with only polynomial costs. In contrast to proposed earlier more complex protocols [2,4,8], the DLCZ scheme is attainable for current experimental technologies. This significant advance has been reached by exploiting the collective enhancement of atom-light interactions provided by optically thick atomic ensembles due to many atoms constructive interference effects [9–13] resulting in collectively enhanced signal-to-noise ratio.

The initial step toward realization of DLCZ protocol is generation of an entanglement between remotely located atomic ensembles. To that end the atomic ensembles with all atoms are prepared initially in the ground state |1⟩ (Fig 1a) are illuminated by a short-pulse weak Raman-pumping laser (referred to as a write laser), that induces a Stokes-photon emission in the transition |3⟩ →|2⟩. Due to coherent coupling of different atoms to the Stokes pulse propagating collinearly with the pumping laser, a collective atomic mode is created in the form of long-lived spin-wave excitations. As a result, a strong correlation between these atomic and Stokes-light modes is produced, while other optical modes are weakly correlated with the collective atomic state and contribute to noise. This significant enhancement of signal-to-noise ratio allows one to generate a pure entanglement between two distinct atomic ensembles by interfering and detecting single Stokes photons emitted from them. The challenging task for this scheme is to demonstrate that the collective atomic state can be separately measured and, hence, the quantum correlation between the atomic and signal-Stokes modes can be revealed. This can be done by first converting the atomic spin excitation into state of the single-mode idler (anti-Stokes) light by applying, after a controllable time delay (Fig.1b), a second laser beam (read laser), and then detecting the idler light again through a single photon detector.

Very recently this program has been successfully accomplished in a number of experiments on generation of nonclassical photon pairs both in samples of cold atoms [14–16] and in room-temperature atomic vapor cells [17,18]. An atomic vapor cell has been used also
in another experiment [19] demonstrating convincingly a strong correlation between the Stokes and anti-Stokes pulses in the regime of large photon number. Meanwhile, a theoretical description of these processes is absent with the exception of some particular results obtained for only the Stokes-photon emission [7] or on the basis of phenomenological approach [14]. Since in the DLCZ scheme we deal only with spontaneous emission, a fully quantum treatment is needed.

In this paper, we develop the quantum theory of creation of atomic memory and generation of nonclassical photon pairs via the DLCZ protocol. In our approach, to exploit the collective enhancement of the atom-light coupling, we consider the interaction between atoms and forward-scattered Stokes and anti-Stokes photons in low-finesse ring cavity [20] with an independent photon emission by individual atoms. The correlation between Stokes and anti-Stokes photons, separated by time interval considerably larger than the excited atomic state lifetime, is established via temporal dynamics of the collective atomic mode. The equation of motion for the latter is derived by adiabatic elimination of cavity modes in the bad-cavity limit. The probability of excitation by the write and read lasers is assumed to be small and is treated in a perturbative way. Our results describe analytically the evolution of quantum correlations, including their dependence on coherent atomic memory lifetime and on time delay between the write and read laser pulses.

For future technological developments, new sources for single-photons are needed, which produce one photon on demand, at a specific time and not at random. A remarkable feature of the DLCZ protocol is the ability to produce not only single-photon pulses, but also any specific multiphoton state on demand via conditional measurement on quantum systems of correlated photon pairs. Compared to other systems of conditional generation of single photons, such as atomic cascades [21,22] and parametric down-conversion [23], this scheme has an advantage of simultaneous control over both the photon number and the spatio-temporal shape of generated pulses that has been recently demonstrated in [15,16,18]. Our theory reproduces the prominent features of observed results including pulse shapes and conditional generation of anti-Stokes single-photon pulse, as well as strong violation of the Cauchy-Schwarz inequality. We believe that the calculations with this simple model may directly be applied to realistic situation of free-space atomic cells. This expectation is supported also by an important observation [24] that the three-dimensional theory confirms the large enhancement of signal-to-noise ratio predicted by simple cavity-QED models [7,11,25].

The outline of the paper is as follows. In section II we derive the master equation for the collective atomic mode by eliminating the degrees of freedom associated with the cavity fields. The mean numbers of the Stokes and anti-Stokes photons inside and emitted from cavity are found in section III. Here we compare our results for photon fluxes with the experimentally observed pulse shapes. In section IV we calculate the photon statistics and cross-correlation between the fields and show a strong violation of Cauchy-Schwarz inequality. In section V we obtain the probability of conditional generation of single anti-Stokes photon and show its dependence on spin-decoherence time. The conclusions are summarized in section VI.

II. DYNAMICAL EQUATION FOR COLLECTIVE ATOMIC MODE

We consider a large ensemble of \( N \) atoms with a level structure shown in Fig.1 and assume that initially all the atoms are prepared by optical pumping in the ground state \( |1\rangle \). The sample in the state \( \Psi_0 \) (Fig.1b) interacts with a weak write pulse with the duration \( T_W \). The write laser beam acts on the transition \( |1\rangle \rightarrow |3\rangle \) with a large detuning \( \Delta_W \) and Rabi frequency \( \Omega_W \) and induces spontaneous emission of a Stokes photon in transition \( |3\rangle \rightarrow |2\rangle \) while flipping an atomic spin into the second ground state \( |2\rangle \). Detection of a forward propagating Stokes photon projects the state of atomic sample onto the nonclassical collective state \( \Psi_1 \) with the symmetric distribution of the flipped spin excitations (see below). The read pulse acting on the transition \( |2\rangle \rightarrow |4\rangle \) with the Rabi frequency \( \Omega_R \) and duration \( T_R \) is applied after time delay \( \tau_d \) and converts the stored spin excitations into the anti-Stokes light. Different schemes can be used for this retrieval process. It is possible to convert the stored spin excitations into an anti-Stokes photon in the electromagnetically induced transparency (EIT [26]) configuration similar to that exploited in previous experiments [12,13] for restoring the classical light pulse. This mechanism has been used in the most of the experiments with DLCZ scheme [14–16,18]. Another way to transfer efficiently a quantum state from atoms to light is off-resonant Raman configuration demonstrated experimentally in [17]. Below, we consider for simplicity the Raman configuration for emission of anti-Stokes photon. Nevertheless, our results describe very well the experimental data obtained with EIT configuration [15,18].

The fields of write and read lasers propagating along the axis of pencil-shape atomic ensemble in z direction are given by

\[
E_{W,R}(t) = E_{W,R} f_{W,R}(t) \exp(i k_w R z - i \omega_{W,R} t),
\]

where \( f_{W,R}(t) \) are the temporal profiles of the pulses.

The positive-frequency parts of forward propagating Stokes and anti-Stokes cavity fields at frequencies \( \omega_1 \) and \( \omega_2 \) are expressed, respectively, in terms of annihilation (creation) operators \( a_1(a_1^\dagger) \) and \( a_2(a_2^\dagger) \) as follows:
where $V$ is a quantization volume, which is assumed to be equal to the volume of the medium.

Owing to the large detunings $\Delta_1 = \omega_3 - \omega_W$ and $\Delta_2 = \omega_4 - \omega_R$ of the write and read lasers from the respective transitions, we can adiabatically eliminate the upper states $|3\rangle$ and $|4\rangle$, which are different in general. Then, under the conditions $(k_W - k_1)L \leq 1$ and $(k_R - k_2)L \leq 1$, where $L$ is the length of the atomic sample, the interaction Hamiltonian for total system in the rotating frame has the form

$$H = \hbar \sum_{i=1}^{N} \left[ G(t) \sigma_{21}^{(i)} a_1^+ - F(t) \sigma_{21}^{(i)} a_2 \right] + h.c. \quad (3)$$

In Eq.(3) the summation is taken over all atoms, $\sigma_{\alpha\beta}^{(i)} = |\alpha\rangle \langle \beta|$ is the atomic spin-flip operator in the basis of the two ground states $|1\rangle$ and $|2\rangle$ for the i-th atom and

$$G(t) = g_s \frac{\Omega_W}{\Delta_W} f_{W}^{1/2}(t), \quad F(t) = g_{AS} \frac{\Omega_R}{\Delta_R} f_{R}^{1/2}(t), \quad (4)$$

where the Rabi frequencies of classical fields are $\Omega_W = \mu_3 E_W / \hbar$ and $\Omega_R = \mu_2 E_R / \hbar$ and

$$g_s = \left( \frac{2\pi\omega_1}{V} \right)^{1/2} \mu_{32} \quad \text{and} \quad g_{AS} = \left( \frac{2\pi\omega_2}{V} \right)^{1/2} \mu_{41} \quad (6)$$

are the atom - quantized fields coupling constants, $\mu_{ij}$ is the dipole matrix element of the transition $|i\rangle \rightarrow |j\rangle$. Note that the Stark shifts of the upper levels $|3\rangle$ and $|4\rangle$ induced by the write and read lasers are included in the frequencies of Stokes and anti-Stokes photons, so that $\omega_1 = \omega_W - \omega_2$ and $\omega_2 = \omega_R + \omega_2$.

In terms of the collective spin operators

$$S^+ = \left( \frac{1}{\sqrt{N}} \right) \sum_{i=1}^{N} \sigma_{21}^{(i)}, \quad S = (S^+)^+ \quad (7)$$

the Hamiltonian (3) is written as

$$H = \hbar \sqrt{N} \left[ G(t) S^+ a_1^+ - F(t) S^+ a_2 \right] + h.c. \quad (8)$$

When all atoms are prepared initially in level $|1\rangle$, the states that are coupled by Hamiltonian (8) are totally symmetric. Particularly, if the atomic ensemble is initially in the state $\Psi_0 = |1_1, 1_2, ..., 1_N\rangle$, then upon emitting one Stokes photon it settles down into symmetric state $\Psi_1 = \frac{1}{\sqrt{N}} \sum_{i=1}^{N} |1_1, 2_i, ..., 1_N\rangle$ (see Fig.1).

The system evolution is described by the master equation for the whole density matrix $\rho$ for the atoms and cavity modes [27]

$$\frac{d\rho}{dt} = -\frac{i}{\hbar} [H, \rho] + \frac{(d\rho)_{rel}}{\hbar} \quad (9)$$

where the second term in right hand side (rhs) accounts for all relaxations in the system. With use of the Liouville operator $L(\rho) = (O^+ O \rho + \rho O^+ O)$ it is written in the form

$$\frac{(d\rho)_{rel}}{\hbar} = \frac{2}{\hbar} \sum_{i=1}^{N} \sum_{\alpha, \beta} a_\alpha k_i L[a_\beta] + \frac{\Gamma_W(t)}{2} \sum_{i=1}^{N} L[\sigma_{21}^{(i)}] \rho$$

$$+ \frac{\Gamma_R(t)}{2} \sum_{i=1}^{N} L[\sigma_{12}^{(i)}] \rho - \gamma_c \sum_{i=1}^{N} I^{(i)} |\rho - \rho(0)\rangle \langle \rho - \rho(0)|. \quad (10)$$

The first term in rhs of this equation, represents the cavity output at the frequencies $\omega_i( i = 1, 2)$ with $2k_i$ being the photon number damping rate for $i$-th mode, while the second and third terms describe spontaneous decay from the upper atomic states $|3\rangle$ and $|4\rangle$ into the ground levels $|2\rangle$ and $|1\rangle$ resulting in optical pumping (OP) to these states during the interaction with the write and read lasers, respectively. The rates of OP are

$$\Gamma_W(t) = \frac{\Omega_W^2}{\Delta_W^2} f_W(t) \gamma_{32}, \quad \Gamma_R(t) = \frac{\Omega_R^2}{\Delta_R^2} f_R(t) \gamma_{41}, \quad (11)$$

where $\gamma_{ij}$ is a partial decay rate of upper level $i$ to the state $j$.

We do not consider here two other channels ($|3\rangle \rightarrow |1\rangle$ and $|4\rangle \rightarrow |2\rangle$) of spontaneous emission, because they do not change the atomic spin distribution. The last term in rhs of Eq.(10) introduces into the model the relaxation of the atomic ground-state coherence at the rate $\gamma_c$, which is supposed to be much smaller compared to the optical coherence damping rate $\gamma$. In this term $f^{(i)} = \sigma_{11}^{(i)} + \sigma_{22}^{(i)}$ represents the unit matrix in the basis of atomic states.

Upon introducing the operators of the atomic populations of the ground states $|1\rangle$ and $|2\rangle$, $S_{\alpha} = \sum_{i=1}^{N} \sigma_{\alpha\alpha}^{(i)}, \ i = 1, 2$, we obtain from Eq.(10) the following equation for average number of the atoms in the state $|2\rangle$:

$$\frac{d}{dt} \langle S_2 \rangle = \left( \frac{d}{dt} + 2k \right) \langle n_{2}^{(in)} \rangle + \Gamma_1(t) N - \gamma_c + \Gamma_2(t) \langle S_2 \rangle \quad (12)$$

where $n_{2}^{(in)} = \langle a_1^+ a_1 \rangle, \ i = 1, 2$, are the mean photon numbers in, respectively, the Stokes and anti-Stokes modes inside the cavity. To simplify, the decay constants of photon numbers, $k_1$ and $k_2$, are taken both equal to
k. Since the average number of output photons $n_i^{(\text{out})}$ is determined by the equations

$$\frac{dn_i^{(\text{out})}}{dt} = 2kn_i^{(\text{in})}, \quad i = 1, 2,$$  \hspace{1cm} (13)

we have

$$\frac{d\langle S_2 \rangle}{dt} = \frac{d}{dt}(n_1^{(\text{tot})} - n_2^{(\text{tot})}) + \Gamma_1(t)N - [\gamma_c + \Gamma_2(t)]\langle S_2 \rangle,$$  \hspace{1cm} (14)

where $n_i^{(\text{tot})} = n_i^{(\text{in})} + n_i^{(\text{out})}$ is the total number of photons in i-th mode.

The physical meaning of this equation is obvious showing that the atomic population in the state $|2\rangle$ is proportional to the photon number difference in Stokes and anti-Stokes modes. Also, it increases due to OP from the state $|1\rangle$, when the atomic sample interacts with the write pulse, and this is described by the term $\Gamma_1 N$. Later on, it decreases at the rate $\Gamma_2$ proportional to the read pulse intensity, as well as due to escape of the atoms from the laser beams area at the rate $\gamma_c$.

We now eliminate the cavity fields adiabatically treating the two-photon interaction terms $G(t)$ and $F(t)$ in the Hamiltonian (8) in a perturbative way. By adiabatic elimination we obtain the equation of motion for the reduced density matrix of the atoms $\rho_a = Tr_c \rho$ in the form

$$\frac{d\rho_a}{dt} = \frac{\alpha(t)}{2} L[S^+]\rho_a + \frac{\beta(t)}{2} L[S]\rho_a + (\frac{d\rho_a}{dt})_{\text{rel}},$$  \hspace{1cm} (15)

where

$$(\frac{d\rho_a}{dt})_{\text{rel}} = \frac{\Gamma_W(t)}{2} \sum_{i=1}^{N} L[\sigma_{2i}^{(1)}]\rho_a + \frac{\Gamma_R(t)}{2} \sum_{i=1}^{N} L[\sigma_{12}^{(i)}]\rho_a - \gamma_c \sum_{i=1}^{N} I^{(i)} [\rho_a - \rho_a(0)]$$  \hspace{1cm} (16)

and

$$\alpha(t) = \frac{2N}{k} G^2(t) = \alpha f_W(t),$$  \hspace{1cm} (17)

$$\beta(t) = \frac{2N}{k} F^2(t) = \beta f_R(t)$$  \hspace{1cm} (18)

are the Stokes gain and anti-Stokes absorption (from the state $|1\rangle$) coefficients, respectively.

This is the central equation of our paper. In deriving Eq.(15), we have assumed only, apart from the weak interaction condition, that the write and read pulses are not overlapped in time. Using Eq.(15), we study in next sections the quantum dynamics of the system including the evolution of collective atomic mode, photon statistics and nonclassical correlations between the photons.

### III. MEAN PHOTON NUMBERS. PULSE SHAPES.

In this section we apply Eq.(15) to find the mean photon numbers in forward-scattered Stokes and anti-Stokes modes inside the cavity and the photon fluxes, given by Eq.(13), as a function of time, as well as the number of spin-wave excitations in the collective atomic mode.

For the overall atomic population in the state $|2\rangle$ we obtain from Eq.(15)

$$\frac{d\langle S_2 \rangle}{dt} = \alpha(t)[N_{sp}(t) + 1] - \beta(t)N_{sp}(t) + \Gamma_1(t)N - [\gamma_c + \Gamma_2(t)]\langle S_2 \rangle,$$  \hspace{1cm} (19)

where $N_{sp} = \langle S^+ S \rangle$ is a number of spin-wave excitations or in other words, a number of the atoms with flipped spin, which form the collective atomic mode. It obeys the equation

$$\frac{dN_{sp}}{dt} = \alpha(t)[N_{sp}(t) + 1] - \beta(t)N_{sp}(t) + \Gamma_1(t) - \Gamma_{\text{tot}}$$  \hspace{1cm} (20)

where $\Gamma_{\text{tot}} = \gamma_c + \Gamma_1(t) + \Gamma_2(t)$.

In obtaining Eqs.(19,20), the commutation relation

$$\langle[S, S^+]\rangle \simeq 1$$  \hspace{1cm} (21)

has been used, which follows from weak interaction condition implying that upon interacting with the write laser, almost all the atoms are maintained in the ground state $|1\rangle$.

The only difference between the Eqs.(19,20) is due to the relaxation terms. Consequently, the total number of the atoms $\langle S_2 \rangle$ in the state $|2\rangle$ is essentially larger than the number of spin-wave excitations, because the former is basically determined by the optical pumping $\Gamma_1 N$ from the ground state $|1\rangle$, whereas the collective atomic mode is generated at the rate $\alpha(t) << \Gamma_1 N$ as a result of coherent interaction with the write laser. At the same time, owing to the fact that the spin coherence and, hence $N_{sp}$, decays at the OP rate of individual atom, the signal-to-noise ratio in Eq.(20) given by $\alpha/\Gamma_1$ and $\beta/\Gamma_2$ is greatly enhanced due to the large factor of the atom number $N$ [7] (see also bellow).

From the comparison of the Eqs.(14) and (19) in the time intervals $0 \leq t \leq T_W$ and $T_2 \leq t \leq T_2 + T_R$, where $T_2 = T_W + \tau_d$ is the instance of read pulse switching on, we immediately find the equations for the mean photon numbers in cavity modes

$$\frac{dn_1^{(\text{in})}}{dt} = \alpha(t)[N_{sp}(t) + 1] - 2kn_1^{(\text{in})},$$  \hspace{1cm} (22)

$$\frac{dn_2^{(\text{in})}}{dt} = \beta(t)N_{sp}(t) - 2kn_2^{(\text{in})},$$  \hspace{1cm} (23)

It is useful at this point to consider numerical estimations. Reasonable parameters are: light wavelength $\lambda = 800 \text{nm}$, $\omega_W \sim \gamma$, $\Omega_R \sim 10\gamma$, $\gamma \sim 10^7 \text{s}^{-1}$, $\gamma_c \sim 10^4 \text{s}^{-1}$,
$L = 1 \div 10cm, V \sim 1cm^3$, and $N \sim 10^{12}$. In the free-space limit, $k = c/L \sim 3 \cdot 10^8s^{-1}$ is the inverse of the propagation time of the pulses through the atomic sample. Then, from Eqs.(17,18) one has $\alpha \sim 10^8s^{-1}$, $\beta \sim 10^8s^{-1}$, and $\Gamma_1 \sim 10^{-5}\gamma$, $\Gamma_2 \sim 10^{-3}\gamma \sim \gamma_c$.

Thus, the signal-to-noise ratio ($\alpha/\Gamma_1$ or $\beta/\Gamma_2$) is about $\sim 10^4$. At the same time, $\alpha, \beta << k$.

From Eqs.(13) the photon fluxes are given by

$$\frac{dn_1^{(out)}}{dt} = \alpha(t)[N_{sp}(t) + 1],$$

$$\frac{dn_2^{(out)}}{dt} = \beta(t)N_{sp}(t).$$

The solution of Eq.(20) with the initial value $N_{sp}(t = 0) = 0$ has the form

$$N_{sp}(t) = \frac{t}{\int_0^t \alpha(t')\exp\{\int_0^{t'} \alpha(\tau) - \beta(\tau) - \Gamma_{tot}(\tau)\}\,d\tau \}.$$  

It is worth noting that if one neglects the relaxations terms in Eq.(20), $n_1^{(out)}(t)$ and $N_{sp}(t)$ obey the same equation during the write pulse. This means that there is an unambiguous correspondence between the number of detected Stokes photons and spin excitations stored in collective atomic mode. In particular, upon interacting with the write laser

$$n_1^{(out)}(T_W) = N_{sp}(T_W).$$

Simple expressions are found for rectangular laser pulses

$$n_1^{(in)}(t) = \frac{\alpha}{2k}e^{\alpha t},$$

$$n_1^{(out)}(t) = e^{\alpha t} - 1, \quad 0 \leq t \leq T_W$$

and

$$n_2^{(in)}(t) = \frac{\beta}{2k}n_1^{(out)}(T_W)e^{-\beta(t-T_2)-\gamma_c t},$$

$$n_2^{(out)}(t) = n_1^{(out)}(T_W) e^{-\gamma_c t}$$

$$\times [1 - e^{-\beta(t-T_2)}], \quad T_2 \leq t \leq T_2 + T_R$$

where $\alpha$ and $\beta$ are defined in Eqs.(17) and (18). It is seen that always $n_1^{(in)} << n_1^{(out)}$. From the Eqs.(30) and (32) it follows also that the numbers of output photons in both modes are the same $n^{(out)}(T_W) = n_2^{(out)}(T_2 + T_R)$ (we replace hereafter, for short, the time argument $T_2 + T_R$ by $T_R$), provided that the intensity of read laser is sufficiently large, $\beta T_R >> 1$, and the time delay between the laser pulses is shorter compared to the spin decoherence time: $\gamma_c T_2 >\gamma_c t_d < 1$.

In Fig.2 we show the Stokes and anti-Stokes pulse shapes calculated by means of the Eqs.(26,27) for the values of parameters given above and for almost rectangular laser pulses with the rise and fall times much shorter than the pulse duration. These results coincide with the theoretical calculations presented in [18] and reproduce very well the experimental data reported in [16,18], although in these experiments the anti-Stokes pulse has been retrieved in EIT configuration. Fig.2a demonstrates the transition from a spontaneous to stimulated emission of Stokes photons with increasing of write pulse intensity. In Fig.2b, the flux of anti-Stokes photons is depicted as a function of time for a fixed number of detected Stokes photons $n_1^{(out)}(T_W) = 3$. The total number of emitted anti-Stokes photons is determined by the areas of the corresponding peaks. It can be shown that in the case of strong read pulse, the stored spin excitations is completely converted into the anti-Stokes photons, i.e. $N_{sp}(T_W) = n_1^{(out)}(T_W) + n_2^{(out)}(T_R)$. This is evident also from Fig.3, where the evolution of spin-wave excitations is shown for different values of read laser intensity with a fixed Raman scattering rate $\alpha(t)$ corresponding to $n_1^{(out)}(T_W) = 3$. Indeed, as it follows from Fig.3, after interaction with the read laser, $N_{sp}(T_R) = 0$ for $\Omega_{R} >> \Omega_{W}$, whereas a residual coherent excitation is preserved in atomic ensemble, if $\Omega_{R} \sim \Omega_{W}$. To demonstrate how the spin decoherence deteriorates the atomic memory, we show in Fig.3b the same calculations for the case of ten times larger decoherence rate $\gamma_c$ and with the same time delay between the write and read laser pulses. In this case the total number of retrieved spin excitations and, hence, of produced anti-Stokes photons is strongly reduced.

IV. PHOTON STATISTICS AND CORRELATIONS. VIOLATION OF CAUCHY-SCHWARZ INEQUALITY.

The nonclassical character of the Stokes and anti-Stokes fields generated in the DLCZ scheme has been experimentally studied in Refs. [14–18] by observing the violation of the Cauchy-Schwarz inequality.

It is well known [28] that two electromagnetic fields, for which a positive true probability distribution exists, satisfy the following Cauchy-Schwarz inequality

$$\left|g^{(12)}\right|^2 \leq g^{(11)}g^{(22)},$$

where $g^{(ii)}$ is the normalized second order auto-correlation functions for i-th field and $g^{(12)}$ is the cross-
correlation between the two fields. They are defined as
\[ g^{(ii)}(t) = \frac{G^{(ii)}(t)}{\langle n_i(t) \rangle} = \frac{\langle a_i^+(t)a_i^+(t)\rangle}{\langle n_i(t) \rangle} \]
\[ g^{(12)}(t_1, t_2) = \frac{G^{(12)}(t_1, t_2)}{\langle n_1(t_1) \rangle \langle n_2(t_2) \rangle} \]
\[ = \frac{\langle a_1^+(t_1)a_2^+(t_2)a_2(t_2)a_1(t_1) \rangle}{\langle n_1(t_1) \rangle \langle n_2(t_2) \rangle} \]
with \( n_i(t) = \langle a_i^+(t)a_i(t) \rangle, \ i = 1, 2 \), being the mean photon numbers.

The inequality (33) is violated for quantized fields. In our case, the correlation functions \( G^{(ij)}(t) \) are easily calculated for the cavity modes. Then, taking into account that in the bad-cavity limit the output fields and cavity modes have obviously the same photon statistics, we apply the obtained results for the detected photons.

Using the Hamiltonian (8), the Heisenberg-Langevin equations for the cavity modes are given by
\[ a_1 = \sqrt{N} \frac{G(t)}{k} S^+ + \int_0^t dt' F_1(t')e^{-k(t-t')}, \] (36)
\[ a_2 = \sqrt{N} \frac{E(t)}{k} S^- + \int_0^t dt' F_2(t')e^{-k(t-t')}, \] (37)
where the noise operators \( F_i(t) \) associated with cavity losses in the Stokes and anti-Stokes modes have the properties [29]
\[ \langle F_i(t) \rangle = \langle F_i(t)F_i(t') \rangle = \langle F_i^+(t)F_i(t') \rangle = 0 \]
\[ \langle F_i(t)F_i^+(t') \rangle = 2k_3\delta \delta(t-t'). \] (38)
The Eqs.(36) and (37) reproduce the solutions (24) and (25) for mean photon numbers \( n_1^{(in)} \) and \( n_2^{(in)} \), if the following conditions:
\[ \langle S(t)F_i(t') \rangle = \langle F_i^+(t')S^+(t) \rangle = 0, \ i = 1, 2 \] (39)
are satisfied for \( t \geq t' \). By using the solution for \( S(t) \) obtained with the Hamiltonian (8) and applying again the properties of Langevin forces (38) it may be proved directly that these correlations must vanish. Moreover, the similar calculations show that this is true also in general case of correlation functions with more than two field operators written in normal order. This allows us, keeping only the first terms in Eqs.(36) and (37), to express the correlation functions \( G(t) \) in terms of the atomic collective spin operators as
\[ G^{(11)}(t) = \left( \frac{\alpha(t)}{2k} \right)^2 \Phi_1(t), \] (40)
\[ G^{(22)}(t) = \left( \frac{\beta(t)}{2k} \right)^2 \Phi_2(t), \]
\[ \Phi_1(t) = \exp \left\{ \int_0^t \left[ 2\alpha(t') - \gamma_c \right] dt' \right\}, \ t \leq T_W \] (44)
\[ \Phi_2(t) = \exp \left\{ - \int_{T_W}^t \left[ 2\beta(t') + \gamma_c \right] dt' \right\}, \ t \geq T_W \] (45)
and have the following simple solutions
\[ \Phi_1(t) = \Phi_1(0) \exp \left\{ \int_0^t \left[ 2\alpha(t') - \gamma_c \right] dt' \right\}, \ t \leq T_W \] (44)
\[ \Phi_2(t) = \Phi_2(T_W) \exp \left\{ - \int_{T_W}^t \left[ 2\beta(t') + \gamma_c \right] dt' \right\}, \ t \geq T_W \] (45)
which, together with \( \Phi_1(T_W) \) from Eq.(44), yields
\[ \Phi_2(T_W) = \langle S^+(T_W)S(T_W) \rangle = \exp \left[ \int_0^{T_W} \alpha(z')dz' \right] - 1 \] (46)
This relation indicates a chaotic nature of spin-wave bosonic excitations just like a similar relation \( \langle a^+a^+a^+a^+ \rangle \)
takes place for a thermal light.

By substituting the Eqs.(40,41,44,45,47) and (24,25) for mean photon numbers \( n_1^{(in)}(T_W) \) and \( n_2^{(in)}(T_W) \) into Eqs.(34), we eventually have
\[ g^{(11)}(t \leq T_W) = 2 \] and \( g^{(22)}(t \geq T_2) = 2 \exp[\gamma_c(t - T_W)] \] (48)
showing that both the Stokes and anti-Stokes modes satisfy Gaussian statistics. It is worth noting that the correlations \( g^{(ii)} \) are independent of time apart from the last factor in \( g^{(22)}(t) \) indicating that an increase of time delay between the laser pulses or of spin-decoherence rate \( \gamma_c \) results in a superchaotic statistics of the anti-Stokes field.

Similarly the cross-correlation function \( G^{(12)}(t_1, t_2) \) may be represented in the form
\[ G^{(12)}(t_1, t_2) = \frac{\alpha(t_1)\beta(t_2)}{2k^2} \Phi_{12}(t_1, t_2); \]
\[ \Phi_{12}(t_1, t_2) = \langle S(t_1)S^+(t_2)S(t_2)S^+(t_1) \rangle \] (49)
where \( t_1 \leq T_W \) and \( t_2 \geq T_2 \). The equation for \( \Phi_{12}(t_1, t_2) \) is deduced from Eq.(20) by applying the quantum regression theorem \([27,30]\) with \( t_2 = t_1 + \tau \), where \( \tau \geq \tau_d \), yielding

\[
\frac{d\Phi_{12}(t_1, t_1 + \tau)}{dt} = -[\beta(t_1 + \tau) + \gamma_c] \phi_{12}(t_1, t_1 + \tau); \tag{50}
\]

the solution of which is

\[
\Phi_{12}(t_1, t_1 + \tau) = \phi_{12}(t_1, t_1) \exp\left[-\int_{t_1}^{t_1+\tau} \beta(t')dt' - \gamma_c\tau\right] \tag{51}\]

Using Eq.(47), \( \Phi_{12}(t_1, t_1) \) is easily transformed to

\[
\Phi_{12}(t_1, t_1) = \langle S(t_1)S^+(t_1) \rangle \left[ 2 \langle S^+(t_1)S(t_1) \rangle + 1 \right] \tag{52}\]

From Eq.(33), the cross-correlation between the two modes is then obtained as

\[
g^{(12)}(t_1, t_2) = 2 + \frac{1}{\langle S^+(t_1)S(t_1) \rangle} \tag{53}\]

and, thus, for the ratio between \( |g^{(12)}|^2 \) and \( g^{(11)} \cdot g^{(22)} \) at \( t_1 = T_W \) and \( t_2 = T_2 + T_R \) we finally have

\[
R = \frac{|g^{(12)}(t_1, t_2)|^2}{g^{(11)}(t_1) g^{(22)}(t_2)} = \left( \frac{1 + 2n_1^{(out)}(T_W)}{2n_1^{(out)}(T_W)} \right)^2 \exp(-\gamma_c\tau_d) \tag{54}\]

where \( \langle S^+(T_W)S(T_W) \rangle \) is replaced by the number \( n_1^{(out)}(T_W) \) of Stokes photons emitted from cavity (see Eq.(29)). Remind that in deriving Eqs.(48) and (54) we have neglected all atomic relaxations during the interaction with laser pulses under assumptions \( \Gamma_{tot} \ll \alpha, \beta \), but we have kept the term \( \gamma_c\tau_d \), which is by no means small and leads to a loss of atomic memory that is manifested by exponential decay of the ratio \( R \).

It follows from Eq.(54) that if the write pulse is sufficiently weak, so that the excitation probability \( p \) in the Stokes channel is less than unity and, hence, \( n_1^{(out)}(T_W) \approx \alpha T_W = p \ll 1 \), the Cauchy-Schwarz inequality is strongly violated by the law \( R \sim 1/p^2 \gg 1 \). In the experiments \([14-17]\) the violation of this inequality has been tested to confirm the non-classical correlations between the Stokes and anti-Stokes pulses and a significant violation has been observed for small \( p \), although, as has been discussed in \([14,15]\), several imperfections limit, in practice, the degree of violation of Cauchy-Schwarz inequality and lead to deviation from the ideal case expressed by Eq.(54).

V. PRODUCING SINGLE PHOTONS VIA CONDITIONAL MEASUREMENT.

The strong non-classical correlation between the Stokes and anti-Stokes pulses in DLCZ scheme can be employed for producing single photons as a result of conditional measurement on correlated photon pairs (see also \([22,23,31]\)), when a detection of one Stokes photon tightly projects the anti-Stokes field into an one-photon state. In order to quantify the confidence level of this procedure we analyze the third-order correlation function \([28]\)

\[
g^{(3)}(t_1, t_2, t_2) = \frac{P_c(t_1, t_2 | 1_1)}{|P_c(1_1 | 1_1)|^2}, \tag{55}\]

which is less unity for quantized fields (it approaches zero in the case of creation of single photons), while \( g^{(3)}(t_1, t_2, t_2) \geq 1 \) for classical fields. In Eq.(55), \( P_c(t_1, t_2 | 1_1) \) and \( P_c(1_1 | 1_1) \) are conditional probabilities for detection of two and one photon from anti-Stokes field, respectively, conditioned upon the detection of one photon in the Stokes channel. Using the known formulae \([28,30]\), the Eq.(55) may be transformed to

\[
g^{(3)}(t_1, t_2, t_2) = \frac{P(1_1)P(1_1, 1_2 | 1_1)}{P(1_1, 1_2)^2}, \tag{56}\]

where the total probabilities are given by

\[
P(1_1) = \langle a_1^+(t)a_1(t) \rangle = n_1^{(in)}(t_1) \tag{57}\]

\[
P(1_1, 1_2) = G^{(12)}(t_1, t_2) \approx \frac{\alpha(t_1) \beta(t_2)}{2k} \frac{\beta(t_2)}{2k} \tag{58}\]

\[
\times \langle S(t_1)S^+(t_1) \rangle \exp\left[-\int_{t_1}^{t_2} \beta(t')dt' - \gamma_c(t_2 - t_1)\right] \tag{59}\]

\[
P(1_1, 1_2, 1_2) = G^{(12)}(t_1, t_2, t_2) \tag{60}\]

\[
= \langle a_1^+(t_1)a_2^+(t_2)a_2(t_2)a_2(t_2)a_1(t_1) \rangle \tag{61}\]

Here we have kept only the second term in \( G^{(12)}(t_1, t_2) \) (see Eqs.(49) and (52)) taking into account the smallness of \( n_1^{(out)}(t_1) = \langle S^+(t_1)S(t_1) \rangle \).

The last correlation function in Eqs.(57) is calculated by the same method employed in previous sections

\[
G^{(12)}(t_1, t_2, t_2) = \frac{\alpha(t_1)}{2k} \frac{\beta(t_2)}{2k} \Phi(t_1, t_2, t_2), \tag{62}\]

\[
\Phi(t_1, t_2, t_2) = \langle S(t_1)S^{+2}(t_2)S^2(t_2)S^+(t_1) \rangle, \tag{63}\]

where \( t_1 \leq T_W, t_2 \geq T_2 \). We obtain the equation for \( \Phi(t_1, t_2, t_2) \) by applying again the quantum regression theorem to the Eq.(43) and find the following solution
\[ \Phi(t_1, t_2, t_2) = \Phi(t_1, t_1, t_1) \exp \left\{ - \int_{t_1}^{t_2} \left[ 2\beta(t') + \gamma_c \right] dt' \right\} \]

where, for \( \Phi(t_1, t_1, t_1) \), by solving the corresponding equation obtained from Eq.(15), we have

\[ \Phi(t_1, t_1, t_1) = 4[n_1^{(out)}(t_1) + 1] \left[ \frac{3}{2} n_1^{(out)}(t_1) + 1 \right] n_1^{(out)}(t_1) \]

Substituting Eqs.(57) and (58) into Eq.(56), we finally get

\[ g^{(3)}(T_W, T_R, T_R) = 4n_1^{(out)}(T_W) \times \left[ \frac{3}{2} n_1^{(out)}(t_W) + 1 \right] \exp(\gamma_c \tau_d) \]

In the regime of very weak Raman excitation, \( n_1^{(out)}(T_W) \approx p \ll 1 \), and provided that \( \gamma_c \tau_d < 1 \) one has \( g^{(3)} \approx 4p \ll 1 \) that corresponds to ideal case of single photon creation.

Experimentally, the conditional generation of single photons in the anti-Stokes pulse has been recently demonstrated with \( g^{(3)} \approx 0.3 \) in an ensemble of cold Cs atoms [15]. The possibility of conditional preparation of Fock states with a given number of photons in optically dense medium of Rb atoms has been reported in Ref. [18].

VI. SUMMARY

In summary, we have developed a theory in the weak pumping limit to describe analytically the non-classical correlations between the photons emitted separately from a medium with macroscopic atomic memory for single-photon fields. First, the spin excitations in atoms are produced in a Raman process induced by the write pulse and are entangled with forward-scattered Stokes photons, and, finally, they are converted by the read pulse into the anti-Stokes photons with an efficiency that depends on the read laser intensity. An important result of our study is that the correlations between the photons are purely determined by only the atomic spin correlations. This allows one to describe the evolution of the system by the dynamical equation for the atoms, instead of solving the underlying equations for quantized pulses. We have derived the master equation for atomic dynamics and have found its analytical solutions for consecutive write and read pulses with time separation longer than the duration of the pulses. The time dependence of photon correlations shows that all quantum effects (violation of Cauchy-Schwarz inequality, conditional generation of single photons, etc) are robustly manifested within the atomic memory lifetime. Otherwise, they exponentially disappear at the atomic spin decoherence rate. To minimize the dissipation we considered the Raman scheme for mapping the atomic spin state onto the anti-Stokes light. Our results obtained in bad-cavity limit are consistent with those observed in single pass scheme with dense atomic media, where for retrieving of the anti-Stokes light the EIT configuration has been used [15,16,18]. From this point of view, the theory we developed may serve as a basic approach for quantum description of storage and retrieval of quantum information, especially when the statistical properties of non-classical pulses are studied.

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FIGURE CAPTIONS

Fig.1. a) Atomic level structure for emission of Stokes and anti-Stokes photons in off-resonant Raman configuration with one photon detunings $\Delta_1$ and $\Delta_2$, respectively. b) Interaction diagram in the case of one Stokes-photon emission, where in initial state $\Psi_0$ all atoms are prepared in the ground state $|1\rangle$. In intermediate stage, the atomic ensemble is in a collective state $\Psi_1$ with symmetric distribution of one atom excitation. c) Time sequence for the write and read pulses having durations $T_W$ and $T_R$. $\tau_d$ is time-delay between the pulses.

Fig.2. a) Stokes photon flux $dn_1^{\text{(out)}}/dt$ as a function of time for different values of write pulse intensity. The curves are labeled with the corresponding values of $\alpha$. The total number of Stokes photons emitted from the cavity are determined by the areas of the corresponding peaks. The duration of write pulse $T_W = 1.6 \mu s$. b) Anti-Stokes photon flux $dn_2^{\text{(out)}}/dt$ for fixed number of output Stokes photons $n_1^{\text{(out)}}(T_W) = 3$ and different values of read laser intensity. The read pulse duration $T_R = 1 \mu s$. The time-delay $\tau_d = 1.4 \mu s$ and $\gamma_c = 0.03 \mu s^{-1}$.

Fig.3. a) Average number of spin-wave excitations $N_{sp} = \langle S^+(t)S(t) \rangle$ as a function of time for fixed number of detected Stokes photons $n_1^{\text{(out)}}(T_W) = 3$ and the values of read laser intensity, as in Fig.2b. $\gamma_c = 0.03 \mu s^{-1}$. b) The same, but $\gamma_c = 0.3 \mu s^{-1}$.
Fig. 1
Fig. 2

a) \[ \frac{dn_1^{(\text{out})}}{dt} \]

- \( 1.16 \text{(µs)}^{-1} \)
- \( 0.874 \text{(µs)}^{-1} \)
- \( 0.6 \text{(µs)}^{-1} \)
- \( \alpha=0.3 \text{(µs)}^{-1} \)

b) \[ \frac{dn_2^{(\text{out})}}{dt} \]

- \( \alpha=0.874 \text{(µs)}^{-1} \)
- \( 10\alpha \)
- \( 5\alpha \)
- \( 1.5\alpha \)

\( \beta=\alpha \)
Fig. 3

(a) \( N_{sp} \)

\[ \alpha = 0.874 \, (\mu s)^{-1} \]

\[ \gamma_c = 0.03 \, (\mu s)^{-1} \]

\[ 10\alpha, 3\alpha, 1.5\alpha, \beta = \alpha \]

(b) \( N_{sp} \)

\[ \gamma_c = 0.3 \, (\mu s)^{-1} \]