Cascade correlation-enhanced Raman scattering in atomic vapors*

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A new Raman process can be used to realize efficient Raman frequency conversion by coherent feedback at low light intensity [Chen B, Zhang K, Bian C L, Qiu C, Yuan C H, Chen L Q, Ou Z Y, and Zhang W P 2013 *Opt. Express* **21**, 10490]. We present a theoretical model to describe this enhanced Raman process, termed as cascade correlation-enhanced Raman scattering, which is a Raman process injected by a seeded light field. It is correlated with the initially prepared atomic spin excitation and driven by the quasi-standing-wave pump fields, and the processes are repeated until the Stokes intensities are saturated. Such an enhanced Raman scattering may find applications in quantum information, nonlinear optics, and optical metrology due to its simplicity.

Keywords: frequency conversions, enhanced Raman scattering, standing-wave pump fields

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1. Introduction

Efficient frequency conversions at low optical powers due to the advent of quantum information science has become important, which has been receiving a lot of attention in recent years.[1–5] At low optical powers, the nonlinear coefficients in most optical materials are usually very small. Traditionally, large optical fields[6] or equivalent long nonlinear medium[7,8] are required to obtain the strong nonlinear response. In the past two decades, it was addressed that the nonlinear conversion can be greatly enhanced using the quantum coherence in atomic medium.[9,10] There are several ways to prepare atomic coherence. One is to use electromagnetically induced transparency (EIT)[9] for the generation of atomic coherence. Jain *et al.*[11] and Merriam *et al.*[12] achieved high frequency conversion efficiencies with the help of an atomic coherence prepared via EIT. Another approach is to prepare an atomic spin wave before the Raman conversion process; the atomic spin wave acts as a seed to the Raman amplification process for enhanced Raman conversion.[13–15] We demonstrated a high Raman conversion of 40% with a lower pump field intensity of 0.1 W/cm^2.[13] Nonlinear conversion efficiency can be enhanced with coherent medium prepared by counter-propagating fields and efficient intrinsic feedback.[16–19] Zibrov *et al.*[17] observed a 4% conversion efficiency with laser power of 300 μW and a spot size of 0.3 mm. However, these schemes need other fields to prepare the atomic spin waves and can only operate in pulse mode.

Recently, we have experimentally demonstrated an efficient Raman conversion scheme with coherent feedback.[20] The experimental setup is simple and the same with usual spontaneous Raman scattering except for a flat mirror. The conversion efficiency of the scheme is as high as 50% for the Stokes field and 30% for the anti-Stokes field with pump field power as low as a few hundreds of a microwatt in both pulsed and continuous wave (CW) modes. By beating two converted fields generated from a common Raman pump field, we observed a narrow line width of 10 kHz, which is determined by the decoherence time of the atomic spin wave in the medium. The mechanism for the efficient conversion is the constructive interference due to the coherent feedback. It relies on the creation of the atomic coherence between the two lower states and the phase correlation between the atomic coherence and converted field in Raman scattering.[21] Recently, we have also proposed a theoretical scheme to enhance the Raman scattering using quantum correlation, termed as correlation-enhanced Raman scattering (CERS).[21] In this scheme, a pump field leads to spontaneous emission of the Stokes field, accompanied with the generation of atomic spin waves. Then this Stokes field used as a seeded signal, with the pump field together, is subsequently input into an atomic ensemble to generate a second Stokes field. Due to the correlation of the seeding Stokes field and atomic spin waves a CERS occurs, which can partly explain the experiment of high efficient Raman conversion, but a system theoretical model is required. In this paper, we present a theoretical model to describe this Raman process, and it can be explained by a cascaded double-CERS. In this model on the one hand the Raman process is driven by the quasi-standing-wave pump fields, and on the other hand the process is repeated and the intensities of Stokes fields are coherent enhancement due to the stronger new initial conditions, finally the high efficient conversion is obtained.

The remaining part of this paper is organized in the fol-

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lowing way. In Section 2 the general model involving spatial propagation and light–atom coupling in Raman scattering driven by the standing wave is given. In Section 3 we explain our results with the above theory. We draw our conclusions in Section 4.

2. Model

In this section, we present a model to describe the process of an efficient Raman conversion scheme with coherent feedback. The schematic diagram of the process considered here is shown in Fig. 1(a). We consider a collection of identical Λ-type hot atoms interacting with laser fields as shown in Fig. 1(b). Two counter-propagating strong pump fields P_1 and P_2 simultaneously couple the ground state $|1\rangle$ to the excited state $|3\rangle$ with one-photon detunings Δ, respectively. Two counter-propagating weak Stokes scattering fields S_1 and S_2 will be generated.

In the electric-dipole and rotating-wave approximations, the Hamiltonian of N identical atoms is given ($\hbar = 1$) as follows:[22,23]

$$\hat{H} = \sum_{j=1}^{N} (\hat{H}_0^j + \hat{H}_{\rm I}^j + \hat{H}_{\rm V}^j), \tag{1}$$

where $\hat{H}_0$ describes the free Hamiltonian of atoms, $\hat{H}_{\rm I}$ describes the interaction of the atoms with the fields, and $\hat{H}_{\rm V}$ describes the interaction Hamiltonian of the atoms with the vacuum electromagnetic field. The individual terms are given by

$$\begin{aligned}
\hat{H}_0^j &= \omega_{31}\hat{\sigma}_{33}^j + \omega_{21}\hat{\sigma}_{22}^j, \\
\hat{H}_{\rm I}^j &= -\big[(\Omega_{P_1} {\rm e}^{-{\rm i}\omega_1 t}{\rm e}^{{\rm i}k_1 z} + \Omega_{P_2}{\rm e}^{-{\rm i}\omega_2 t}{\rm e}^{-{\rm i}k_2 z})\hat{\sigma}_{31}^j \\
&\quad + (g_1\hat{\mathscr{E}}_{S_1}{\rm e}^{-{\rm i}\omega_3 t}{\rm e}^{{\rm i}k_3 z} + g_2\hat{\mathscr{E}}_{S_2}{\rm e}^{-{\rm i}\omega_4 t}{\rm e}^{-{\rm i}k_4 z})\hat{\sigma}_{32}^j + {\rm H.c}\big], \\
\hat{H}_{\rm V}^j &= -\sum_k \hat{b}_k(g_{1k}\hat{\sigma}_{31}^j + g_{2k}\hat{\sigma}_{32}^j + {\rm H.c}),
\end{aligned} \tag{2}$$

where $\hat{\sigma}_{kl}^j = |k\rangle_{jj}\langle l|$ $(k,l=1,2,3)$, $\Omega_i = \mu_{31}E_i/2\hbar$ $(i = P_1, P_2)$ are the half Rabi frequency of pump fields, and g_i $(i=1,2)$ are the coupling strength. $\hat{b}_k$ are boson annihilation operators for the bath and g_i $(i = 1k, 2k)$ are the coupling strengths of the atom with the vacuum electromagnetic fields.

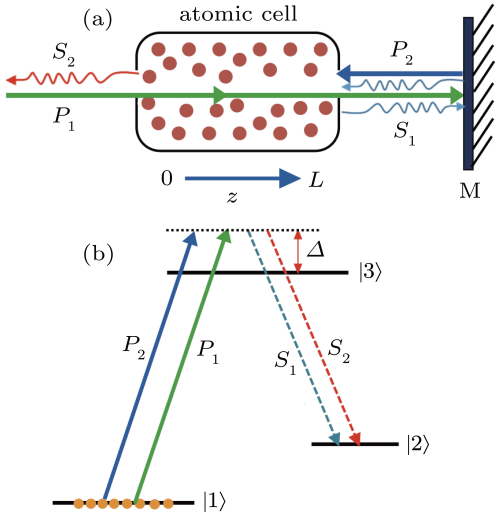


Fig. 1. (color online) (a) The schematic diagram of the experiment. (b) The schematic diagram of Λ-type three-level atoms. P_1, P_2: pump fields; S_1, S_2: Stokes fields; Δ: detuning; M: mirror.

We define continuum atomic operators $\hat{\sigma}_{\mu\nu}$ by summing over the individual atoms in a small volume V, and introduce slowly varying atomic operators $\tilde{\sigma}_{\mu\nu}$: $\tilde{\sigma}_{31} = \hat{\sigma}_{31}{\rm e}^{-{\rm i}\omega_2 t}$, $\tilde{\sigma}_{23} = \hat{\sigma}_{23}{\rm e}^{{\rm i}\omega_4 t}$, $\tilde{\sigma}_{21} = \hat{\sigma}_{21}{\rm e}^{{\rm i}(\omega_4-\omega_2)t}$. Then the equations of motion for the atomic operators in the Heisenberg picture are

$$\partial_t\tilde{\sigma}_{21} = -\gamma_{21}\tilde{\sigma}_{21} + {\rm i}\big(\Omega_{P_1}^*{\rm e}^{-{\rm i}k_1 z} + \Omega_{P_2}^*{\rm e}^{{\rm i}k_2 z}\big)\tilde{\sigma}_{23} - {\rm i}\big(g_1\hat{\mathscr{E}}_{S_1}{\rm e}^{{\rm i}k_3 z} + g_2\hat{\mathscr{E}}_{S_2}{\rm e}^{-{\rm i}k_4 z}\big)\tilde{\sigma}_{31} + \hat{F}_{21}, \tag{3}$$

$$\partial_t\tilde{\sigma}_{31} = -(\gamma_{31} + {\rm i}\Delta)\tilde{\sigma}_{31} - {\rm i}\big(g_1\hat{\mathscr{E}}_{S_1}^\dagger{\rm e}^{-{\rm i}k_3 z} + g_2\hat{\mathscr{E}}_{S_2}^\dagger{\rm e}^{{\rm i}k_4 z}\big)\tilde{\sigma}_{21} - {\rm i}\big(\Omega_{P_1}^*{\rm e}^{-{\rm i}k_1 z} + \Omega_{P_2}^*{\rm e}^{{\rm i}k_2 z}\big)(\hat{\sigma}_{11} - \hat{\sigma}_{33}) + \hat{F}_{31}, \tag{4}$$

$$\partial_t\tilde{\sigma}_{23} = -(\gamma_{23} - {\rm i}\Delta)\tilde{\sigma}_{23} + {\rm i}\big(\Omega_{P_1}{\rm e}^{{\rm i}k_1 z} + \Omega_{P_2}{\rm e}^{-{\rm i}k_2 z}\big)\tilde{\sigma}_{21} + {\rm i}\big(g_1\hat{\mathscr{E}}_{S_1}{\rm e}^{{\rm i}k_3 z} + g_2\hat{\mathscr{E}}_{S_2}{\rm e}^{-{\rm i}k_4 z}\big)(\hat{\sigma}_{22} - \hat{\sigma}_{33}) + \hat{F}_{23}, \tag{5}$$

where $\hat{F}_{\mu\nu}$ are the Langevin noise operators and their detailed expressions are given in Ref. [23]. Due to large detuning $\Delta \gg \Omega_{P_1}$, Ω_{P_2}, γ_{23}, γ_{31}, we obtain

$$\tilde{\sigma}_{23} = -\left(1 + {\rm i}\frac{\gamma_{23}}{\Delta}\right)\left\{\left[\frac{\Omega_{P_1}}{\Delta}{\rm e}^{{\rm i}(k_1-k_3)z}\tilde{\sigma}_{21} + \frac{g_1}{\Delta}\hat{\mathscr{E}}_{S_1}\hat{\sigma}_{22}\right]{\rm e}^{{\rm i}k_3 z} + \left[\frac{\Omega_{P_2}}{\Delta}{\rm e}^{-{\rm i}(k_2-k_4)z}\tilde{\sigma}_{21} + \frac{g_2}{\Delta}\hat{\mathscr{E}}_{S_2}\hat{\sigma}_{22}\right]{\rm e}^{-{\rm i}k_4 z}\right\}, \tag{6}$$

$$\tilde{\sigma}_{31} = -\left(1 + {\rm i}\frac{\gamma_{31}}{\Delta}\right)\left[\left(\frac{\Omega_{P_1}^*}{\Delta}{\rm e}^{-{\rm i}k_1 z} + \frac{\Omega_{P_2}^*}{\Delta}{\rm e}^{{\rm i}k_2 z}\right)(\hat{\sigma}_{11} - \hat{\sigma}_{33}) + \frac{g_1}{\Delta}\hat{\mathscr{E}}_{S_1}^\dagger{\rm e}^{-{\rm i}k_3 z}\tilde{\sigma}_{21} + \frac{g_2}{\Delta}\hat{\mathscr{E}}_{S_2}^\dagger{\rm e}^{{\rm i}k_4 z}\tilde{\sigma}_{21}\right] - {\rm i}\frac{\hat{F}_{31}}{\Delta}. \tag{7}$$

Substituting $\tilde{\sigma}_{23}$ and $\tilde{\sigma}_{31}$ into $\tilde{\sigma}_{21}$, and making a secular approximation, i.e., collecting the terms with the similar oscillatory terms and neglecting the fast oscillating contributions, equation (3) can be written as

$$\begin{aligned}
\partial_t\tilde{\sigma}_{21} \simeq &-\left[\gamma_{21} - {\rm i}\frac{\Omega^2}{\Delta}\left(1 + {\rm i}\frac{\gamma_{23}}{\Delta}\right) + \frac{1}{\gamma_{31} + {\rm i}\Delta}(g_1^2\hat{\mathscr{E}}_{S_1}\hat{\mathscr{E}}_{S_1}^\dagger + g_2^2\hat{\mathscr{E}}_{S_2}\hat{\mathscr{E}}_{S_2}^\dagger)\right]\tilde{\sigma}_{21} \\
&- \frac{1}{\gamma_{31} + {\rm i}\Delta}(g_1\hat{\mathscr{E}}_{S_1}\Omega_{P_1}^*{\rm e}^{-{\rm i}\delta k z} + g_2\hat{\mathscr{E}}_{S_2}\Omega_{P_2}^*{\rm e}^{{\rm i}\delta k z})\hat{\sigma}_{11} - {\rm i}\left(1 + {\rm i}\frac{\gamma_{23}}{\Delta}\right)\left[\frac{g_1\Omega_{P_1}^*}{\Delta}\hat{\mathscr{E}}_{S_1}{\rm e}^{-{\rm i}\delta k z} + \frac{g_2\Omega_{P_2}^*}{\Delta}\hat{\mathscr{E}}_{S_2}{\rm e}^{{\rm i}\delta k z}\right. \\
&\left. - \left(\frac{g_1^*\Omega_{P_1}}{\Delta}\hat{\mathscr{E}}_{S_1}{\rm e}^{-{\rm i}\delta k z} + \frac{g_2\Omega_{P_2}^*}{\Delta}\hat{\mathscr{E}}_{S_2}{\rm e}^{{\rm i}\delta k z}\right)\right]\hat{\sigma}_{22} + \hat{F}_{21},
\end{aligned} \tag{8}$$

where $\delta k = k_1 - k_3 = k_2 - k_4$, $\Omega^2 = |\Omega_{P_1}|^2 + |\Omega_{P_2}|^2$. The above equations suggest the decomposition of the optical coherence in two counter-propagating components $\tilde{\sigma}_{23} = \tilde{\sigma}_{23}^{+}\mathrm{e}^{\mathrm{i}k_3 z} + \tilde{\sigma}_{23}^{-}\mathrm{e}^{-\mathrm{i}k_4 z}$. Then the coherence $\tilde{\sigma}_{23}^{\pm}$ is written as

$$\tilde{\sigma}_{23}^{+} = -\left(1+\mathrm{i}\frac{\gamma_{23}}{\Delta}\right)\left[\frac{\Omega_{P_1}}{\Delta}\mathrm{e}^{\mathrm{i}(k_1-k_3)z}\tilde{\sigma}_{21} + \frac{g_1}{\Delta}\hat{\mathscr{E}}_{S_1}\hat{\sigma}_{22}\right], \quad (9)$$

$$\tilde{\sigma}_{23}^{-} = -\left(1+\mathrm{i}\frac{\gamma_{23}}{\Delta}\right)\left[\frac{\Omega_{P_2}}{\Delta}\mathrm{e}^{-\mathrm{i}(k_2-k_4)z}\tilde{\sigma}_{21} + \frac{g_2}{\Delta}\hat{\mathscr{E}}_{S_2}\hat{\sigma}_{22}\right]. \quad (10)$$

Using the slowly varying envelope approximation, we obtain the following propagation equations for the quantum field operators:

$$\left(\frac{\partial}{\partial t} + c\frac{\partial}{\partial z}\right)\hat{\mathscr{E}}_{S_1}(z,t) = \mathrm{i}g_1 N\tilde{\sigma}_{23}^{+}, \quad (11)$$

$$\left(\frac{\partial}{\partial t} - c\frac{\partial}{\partial z}\right)\hat{\mathscr{E}}_{S_2}(z,t) = \mathrm{i}g_2 N\tilde{\sigma}_{23}^{-}. \quad (12)$$

If the atom never departs significantly from its initial state $|1\rangle$, then $\hat{\sigma}_{11} - \hat{\sigma}_{33}$ may be replaced by the unit operator. The propagating quantized Stokes fields $\hat{\mathscr{E}}_{S_1}$ and $\hat{\mathscr{E}}_{S_2}$ obey the equation of motion

$$\left(\frac{\partial}{\partial t} + c\frac{\partial}{\partial z}\right)\hat{\mathscr{E}}_{S_1}(z,t) = \mathrm{i}g_1 N\frac{\Omega_{P_1}}{\Delta}\mathrm{e}^{\mathrm{i}\delta kz}\tilde{\sigma}_{21}, \quad (13)$$

$$\left(\frac{\partial}{\partial t} - c\frac{\partial}{\partial z}\right)\hat{\mathscr{E}}_{S_2}(z,t) = \mathrm{i}g_2 N\frac{\Omega_{P_2}}{\Delta}\mathrm{e}^{-\mathrm{i}\delta kz}\tilde{\sigma}_{21}, \quad (14)$$

$$\partial_t\tilde{\sigma}_{21} = -\left[\gamma_{21} + \frac{\Omega^2}{\Delta^2}\gamma_{23} - \mathrm{i}\frac{\Omega^2}{\Delta}\right]\tilde{\sigma}_{21} + \mathrm{i}\left(g_1\frac{\Omega_{P_1}^*}{\Delta}\mathrm{e}^{-\mathrm{i}\delta kz}\hat{\mathscr{E}}_{S_1} + g_2\frac{\Omega_{P_2}^*}{\Delta}\mathrm{e}^{\mathrm{i}\delta kz}\hat{\mathscr{E}}_{S_2}\right) + \hat{F}_{21}. \quad (15)$$

Introducing $\tilde{\mathscr{E}}_{S_1}(z,t) = \hat{\mathscr{E}}_{S_1}(z,t)$, $\tilde{\mathscr{E}}_{S_2}(z,t) = \hat{\mathscr{E}}_{S_2}(z,t)\mathrm{e}^{\mathrm{i}\delta kz}$, $\hat{S}_a^{\dagger}(z,t) = \sqrt{N}\tilde{\sigma}_{21}\mathrm{e}^{\mathrm{i}\delta kz}$, $\chi_1(t) = g_1\sqrt{N}\Omega_{P_1}/\Delta$, and $\chi_2(t) = g_2\sqrt{N}\Omega_{P_2}/\Delta$, the propagating quantized Stokes fields obey the following equations of motion

$$\begin{aligned}
&\left(\frac{\partial}{\partial t} + c\frac{\partial}{\partial z}\right)\tilde{\mathscr{E}}_{S_1}(z,t) = \mathrm{i}\chi_1(t)\hat{S}_a^{\dagger}(z,t),\\
&\left(\frac{\partial}{\partial t} - c\frac{\partial}{\partial z}\right)\tilde{\mathscr{E}}_{S_2}(z,t) = \mathrm{i}2\delta kc\tilde{\mathscr{E}}_{S_2}(z,t) + i\chi_2(t)\hat{S}_a^{\dagger}(z,t),\\
&\partial_t\hat{S}_a^{\dagger}(z,t) = -\Gamma\hat{S}_a^{\dagger}(z,t) + \mathrm{i}[\chi_1^*(t)\tilde{\mathscr{E}}_{S_1}(z,t) + \chi_2^*(t)\tilde{\mathscr{E}}_{S_2}(z,t)] + \hat{F}_S^{\dagger}(z,t),
\end{aligned} \quad (16)$$

where $\Gamma = \gamma_S - \mathrm{i}\Omega^2/\Delta$, $\gamma_S = \gamma_{21} + \gamma_{23}\Omega^2/\Delta^2$, and

$$\langle\hat{F}_S(z,t)\hat{F}_S^{\dagger}(z',t')\rangle = 2\gamma_S L\delta(z-z')\delta(t-t').$$

These equations show that the counter-propagating pump fields Ω_{P_1} and Ω_{P_2} induce a coupling between $\tilde{\mathscr{E}}_{S_1}$ and $\tilde{\mathscr{E}}_{S_2}$, mediated through the spin coherence $\hat{S}_a$.

3. Analytic solution

In this section, we use the Laplace transform technique to solve Eq. (16). Using the moving frame $t' = t - z/c$, $z' = z$, then equation (16) can be rewritten as

$$\begin{aligned}
&\partial_{z'}\hat{\mathscr{E}}_{S_1}(z',t') = \mathrm{i}\chi_1(t')/c\hat{S}_a^{\dagger}(z',t'),\\
&(2\partial_{t'} - c\partial_{z'})\tilde{\mathscr{E}}_{S_2}(z',t') = \mathrm{i}2\delta kc\tilde{\mathscr{E}}_{S_2}(z',t') + \mathrm{i}\chi_2(t')\hat{S}_a^{\dagger}(z',t'),\\
&\partial_{t'}\hat{S}_a^{\dagger}(z',t') = -\Gamma\hat{S}_a^{\dagger}(z',t') + \mathrm{i}[\chi_1^*(t')\tilde{\mathscr{E}}_{S_1}(z',t') + \chi_2^*(t')\tilde{\mathscr{E}}_{S_2}(z',t')] + \hat{F}_S^{\dagger}(z',t').
\end{aligned} \quad (17)$$

To solve the above equations of motions, we first make a Laplace transformation to the Stokes field equations ($\partial_{z'} \to s$)

$$\hat{\mathscr{E}}_{S_1}(s,t') = \frac{1}{s}\left[\hat{\mathscr{E}}_{S_1}(0,t') + \mathrm{i}\frac{\chi_1(t')}{c}\hat{S}_a^{\dagger}(s,t')\right], \quad (18)$$

$$\partial_{t'}\tilde{\mathscr{E}}_{S_2}(s,t') = \frac{1}{2}c(s+2\mathrm{i}\delta k)\tilde{\mathscr{E}}_{S_2}(s,t') + \frac{\mathrm{i}}{2}\chi_2(t')\hat{S}_a^{\dagger}(s,t') - \frac{c}{2}\tilde{\mathscr{E}}_{S_2}(s_L,t'), \quad (19)$$

$$\partial_{t'}\hat{S}_a^{\dagger}(s,t') = -\left[\Gamma - \mathrm{i}\frac{|\chi_1(t')|^2}{cs}\right]\hat{S}_a^{\dagger}(s,t') + \mathrm{i}\chi_2^*(t')\tilde{\mathscr{E}}_{S_2}(s,t') + \frac{\mathrm{i}\chi_1^*(t')}{s}\hat{\mathscr{E}}_{S_1}(0,t') + \hat{F}_S^{\dagger}(s,t'). \quad (20)$$

The initial condition of Stokes field S_2 is $\tilde{\mathscr{E}}_{S_2}(s_L,t')$ instead of $\tilde{\mathscr{E}}_{S_2}(0,t')$ due to the Stokes field S_2 having backward propagation.

For analyzing conveniently, we assume the write fields intensity $[\Omega_{P_1}(t') = \Omega_{P_1}\theta(t')]$ being constant and real, after being switched on at $t' = 0$. Then the coupled equations (19) and (20) are written as

$$\partial_{t'}\tilde{\mathscr{E}}_{S_2}(s,t') = \frac{1}{2}c(s+2\mathrm{i}\delta k)\tilde{\mathscr{E}}_{S_2}(s,t') + \frac{\mathrm{i}}{2}\chi_2\hat{S}_a^{\dagger}(s,t') - \frac{c}{2}\tilde{\mathscr{E}}_{S_2}(s_L,t'), \quad (21)$$

$$\partial_{t'}\hat{S}_a^{\dagger}(s,t') = -\left[\Gamma - \mathrm{i}\frac{|\chi_1|^2}{cs}\right]\hat{S}_a^{\dagger}(s,t') + \mathrm{i}\chi_2^*\tilde{\mathscr{E}}_{S_2}(s,t') + \frac{\mathrm{i}\chi_1^*}{s}\hat{\mathscr{E}}_{S_1}(0,t') + \hat{F}_S^{\dagger}(s,t'), \quad (22)$$

and the solutions of them are given by

$$\tilde{\mathscr{E}}_{S_2}(s,t') = \frac{\chi_2}{2\mathrm{i}\Lambda}\left\{(\nu_-\mathrm{e}^{-\lambda_+ t'} - \nu_+\mathrm{e}^{-\lambda_- t'})\tilde{\mathscr{E}}_{S_2}(s,0) + (-\mathrm{e}^{-\lambda_+ t'} + \mathrm{e}^{-\lambda_- t'})\hat{S}_a^{\dagger}(s,0) + \int_0^{t'}\mathrm{d}t''\left[-\frac{c}{2}(\nu_-\mathrm{e}^{-\lambda_+(t'-t'')} - \nu_+\mathrm{e}^{-\lambda_-(t'-t'')})\tilde{\mathscr{E}}_{S_2}(s_L,t'')\right.\right.$$

$$-(\mathrm{e}^{-\lambda_+(t'-t'')}-\mathrm{e}^{-\lambda_-(t'-t'')})\left(\frac{i}{s}\chi_1^*\hat{\mathscr{E}}_{S_1}(0,t'')+\hat{F}_S^\dagger(s,t'')\right)\Big]\Big\}, \tag{23}$$

$$\hat{S}_a^\dagger(s,t')=\frac{\chi_2}{2\mathrm{i}\Lambda}\Big\{\nu_+\nu_-(\mathrm{e}^{-\lambda_+t'}-\mathrm{e}^{-\lambda_-t'})\tilde{\mathscr{E}}_{S_2}(s,0)$$
$$+(-\nu_+\mathrm{e}^{-\lambda_+t'}+\nu_-\mathrm{e}^{-\lambda_-t'})\hat{S}_a^\dagger(s,0)+\int_0^{t'}\mathrm{d}t''\Big[-\frac{c}{2}\nu_+\nu_-(\mathrm{e}^{-\lambda_+(t'-t'')}-\mathrm{e}^{-\lambda_-(t'-t'')})\tilde{\mathscr{E}}_{S_2}(s_L,t'')$$
$$-(\nu_+\mathrm{e}^{-\lambda_+(t'-t'')}-\nu_-\mathrm{e}^{-\lambda_-(t'-t'')})\left(\frac{\mathrm{i}}{s}\chi_1^*\hat{\mathscr{E}}_{S_1}(0,t'')+\hat{F}_S^\dagger(s,t'')\right)\Big]\Big\}, \tag{24}$$

where

$$\lambda_\pm=-\frac{1}{2}\left[\left(\Gamma-\frac{\mathrm{i}|\chi_1|^2}{cs}\right)-\left(\frac{1}{2}cs+\mathrm{i}\delta kc\right)\mp\Lambda\right],\quad \Lambda=\sqrt{\left[\left(\Gamma-\frac{\mathrm{i}|\chi_1|^2}{cs}\right)+\left(\frac{1}{2}cs+\mathrm{i}\delta kc\right)\right]^2-2|\chi_2|^2},$$
$$\nu_+=\frac{\mathrm{i}}{\chi_2}\left[\left(\Gamma-\frac{\mathrm{i}|\chi_1|^2}{cs}\right)+\frac{1}{2}c(s+2\mathrm{i}\delta k)-\Lambda\right],\quad \nu_-=\frac{\mathrm{i}}{\chi_2}\left[\left(\Gamma-\frac{\mathrm{i}|\chi_1|^2}{cs}\right)+\frac{1}{2}c(s+2\mathrm{i}\delta k)+\Lambda\right]. \tag{25}$$

Then substituting Eq. (24) into Eq. (18), we obtain the solution of Stokes field S_1, which is given by

$$\hat{\mathscr{E}}_{S_1}(s,t')=\frac{\chi_1\chi_2}{2c\Lambda}\Big\{\nu_+\nu_-(\mathrm{e}^{-\lambda_+t'}-\mathrm{e}^{-\lambda_-t'})\tilde{\mathscr{E}}_{S_2}(s,0)+(-\nu_+\mathrm{e}^{-\lambda_+t'}+\nu_-\mathrm{e}^{-\lambda_-t'})\hat{S}_a^\dagger(s,0)$$
$$+\int_0^{t'}\mathrm{d}t''\Big[-\frac{c}{2}\nu_+\nu_-(\mathrm{e}^{-\lambda_+(t'-t'')}-\mathrm{e}^{-\lambda_-(t'-t'')})\tilde{\mathscr{E}}_{S_2}(s_L,t'')-(\nu_+\mathrm{e}^{-\lambda_+(t'-t'')}-\nu_-\mathrm{e}^{-\lambda_-(t'-t'')})$$
$$\times\left(\frac{\mathrm{i}}{s}\chi_1^*\hat{\mathscr{E}}_{S_1}(0,t'')+\hat{F}_S^\dagger(s,t'')\right)\Big]\Big\}+\frac{1}{s}\hat{\mathscr{E}}_{S_1}(0,t''). \tag{26}$$

In order to explain the process of efficient Raman conversion scheme,[20] we calculate the intensities of Stokes field S_1 and S_2. Different initial conditions lead to different output intensities, where the process is described in Fig. 2. The pump field P_1 leads to spontaneous emission of the Stokes field S_1, accompanying with the generation of atomic spin waves S_{a1} shown in Fig. 2(b). The atomic spin wave S_{a1} stays in the cell, the S_1 and P_1 fields propagate out together and both are reflected back to the atomic medium by a flat mirror M (see Fig. 2(c)). If only P_2 is present, the subsequent Stokes field S_2 in the backward direction will be stimulated by the reflected S_1 and enhanced by the previously produced atomic spin wave at the same time, which was studied by us and we call CERS.[21] But in Fig. 2(d) the difference is that the initial correlation condition simultaneously leads to two enhanced Stokes fields S_1 and S_2 generation. This process is double CERS, which is also obtained from Eq. (16). Figures 2(a)–2(d) form the first cycle and figures 2(e)–2(f) form the second cycle. Here the cycles are repeated constantly in a period, and the intensity of Stokes field S_2 is coherent enhancement until it is saturated. Therefore, we term as the cascade CERS. Next we present the expression of the intensities under different conditions.

In the first cycle, for Stokes field S_1 no initial Stokes field is externally incident on the ensemble and no initial spin wave is written into the ensemble (see Fig. 2(a)), then the initial conditions are given by

$$\langle\hat{\mathscr{E}}_{S_1}^\dagger(0,t')\hat{\mathscr{E}}_{S_1}(0,t')\rangle=0,\quad \langle\tilde{\mathscr{E}}_{S_2}^\dagger(s_L,t')\hat{\mathscr{E}}_{S_2}(s_L,t')\rangle=0,$$
$$\langle\hat{S}_a^\dagger(s,0)\hat{S}_a(s,0)\rangle=0,\quad \langle\tilde{\mathscr{E}}_{S_2}^\dagger(s,0)\tilde{\mathscr{E}}_{S_2}(s,0)\rangle=0. \tag{27}$$

The corresponding intensity is worked by

$$I_{S_1}^{\mathrm{spon}}=\frac{|\chi_1\chi_2|^2}{4c^2|\Lambda|^2}\Big\{(\nu_+^*\mathrm{e}^{-\lambda_+^*t'}-\nu_-^*e^{-\lambda_-^*t'})(\nu_+\mathrm{e}^{-\lambda_+t'}$$
$$-\nu_-\mathrm{e}^{-\lambda_-t'})L+\int_0^{t'}\int_0^{t'}\mathrm{d}t''\mathrm{d}t'''(\nu_+^*\mathrm{e}^{-\lambda_+^*(t'-t'')}$$
$$-\nu_-^*\mathrm{e}^{-\lambda_-^*(t'-t'')})(\nu_+\mathrm{e}^{-\lambda_+(t'-t''')}-\nu_-\mathrm{e}^{-\lambda_-(t'-t''')})$$
$$\times\langle\hat{F}_S(s,t'')\hat{F}_S^\dagger(s,t''')\rangle\Big\}, \tag{28}$$

which had been studied by Raymer *et al.*[22] The Stokes field S_1 is generated from the vacuum and the atomic ground state, which is shown in Fig. 2(b).

Subsequently, the atomic spin wave S_a stays in the cell, and the S_1 and P_1 fields propagate out together and both are reflected back to the atomic medium by a flat mirror M, where the reflected P_1 is denoted as P_2. As shown in Fig. 2(c), the Stokes field S_1 with its correlated atomic spin wave $\hat{S}_a$ is employed as initial seeding, and the atomic emsemble is pumped by the standing-wave formed by P_1 and P_2. Then two new enhanced Stokes fields S_1 and S_2 are generated as shown in Fig. 2(d). For Stokes field S_2, the initial conditions are given by

$$\langle\hat{\mathscr{E}}_{S_1}^\dagger(0,t')\hat{\mathscr{E}}_{S_1}(0,t')\rangle=0,\quad \langle\tilde{\mathscr{E}}_{S_2}^\dagger(s_L,t')\hat{\mathscr{E}}_{S_2}(s_L,t')\rangle\neq0,$$
$$\langle\hat{S}_a^\dagger(s,0)\hat{S}_a(s,0)\rangle\neq0,\quad \langle\tilde{\mathscr{E}}_{S_2}^\dagger(s,0)\tilde{\mathscr{E}}_{S_2}(s,0)\rangle=0, \tag{29}$$

and

$$\langle\tilde{\mathscr{E}}^{\dagger}_{S_2}(s_L,t')\hat{S}^{\dagger}_a(s,0)\rangle \neq 0, \tag{30}$$

where the light–atom correlation will play an important role due to the above relation. The corresponding intensity is

$$I_{S_2} = I^{\rm spon}_{S_2} + I^{\rm seed}_{S_2} + I^{\rm Sa}_{S_2} + I^{\rm seed-Sa}_{S_2}, \tag{31}$$

where

$$I^{\rm spon}_{S_2} = \frac{|\chi_2|^2}{4|\Lambda|^2}\Big\{L(\mathrm{e}^{-\lambda^*_- t'} - \mathrm{e}^{-\lambda^*_+ t'})(\mathrm{e}^{-\lambda_- t'} - \mathrm{e}^{-\lambda_+ t'}) + \int_0^{t'}\int_0^{t'} \mathrm{d}t''\,\mathrm{d}t'''(\mathrm{e}^{-\lambda^*_+(t'-t'')} - \mathrm{e}^{-\lambda^*_-(t'-t'')})(\mathrm{e}^{-\lambda_+(t'-t''')} - \mathrm{e}^{-\lambda_-(t'-t''')})$$
$$\times \langle \hat{F}_S(s,t'')\hat{F}^{\dagger}_S(s,t''')\rangle\Big\}, \tag{32}$$

$$I^{\rm seed}_{S_2} = \frac{|\chi_2|^2}{4|\Lambda|^2}\frac{c^2}{4}\int_0^{t'}\int_0^{t'} \mathrm{d}t''\,\mathrm{d}t'''(\nu^*_- \mathrm{e}^{-\lambda^*_+(t'-t'')} - \nu^*_+ \mathrm{e}^{-\lambda^*_-(t'-t'')})(\nu_- \mathrm{e}^{-\lambda_+(t'-t''')} - \nu_+ \mathrm{e}^{-\lambda_-(t'-t''')})$$
$$\times \langle\tilde{\mathscr{E}}^{\dagger}_{S_2}(s_L,t'')\tilde{\mathscr{E}}_{S_2}(s_L,t''')\rangle, \tag{33}$$

$$I^{\rm Sa}_{S_2} = \frac{|\chi_2|^2}{4|\Lambda|^2}\Big\{(\mathrm{e}^{-\lambda^*_- t'} - \mathrm{e}^{-\lambda^*_+ t'})(\mathrm{e}^{-\lambda_- t'} - \mathrm{e}^{-\lambda_+ t'})\langle\hat{S}_a(s,0)\hat{S}^{\dagger}_a(s,0)\rangle, \tag{34}$$

$$I^{\rm seed-Sa}_{S_2} = \frac{|\chi_2|^2}{4|\Lambda|^2}\int_0^{t'} \mathrm{d}t''\left[\frac{c}{2}(\nu_- \mathrm{e}^{-\lambda_+(t'-t'')} - \nu_+ \mathrm{e}^{-\lambda_-(t'-t'')})(\mathrm{e}^{-\lambda^*_+ t'} - \mathrm{e}^{-\lambda^*_- t'})\langle\tilde{\mathscr{E}}_{S_2}(s_L,t'')\hat{S}_a(s,0)\rangle + \mathrm{H.c.}\right]. \tag{35}$$

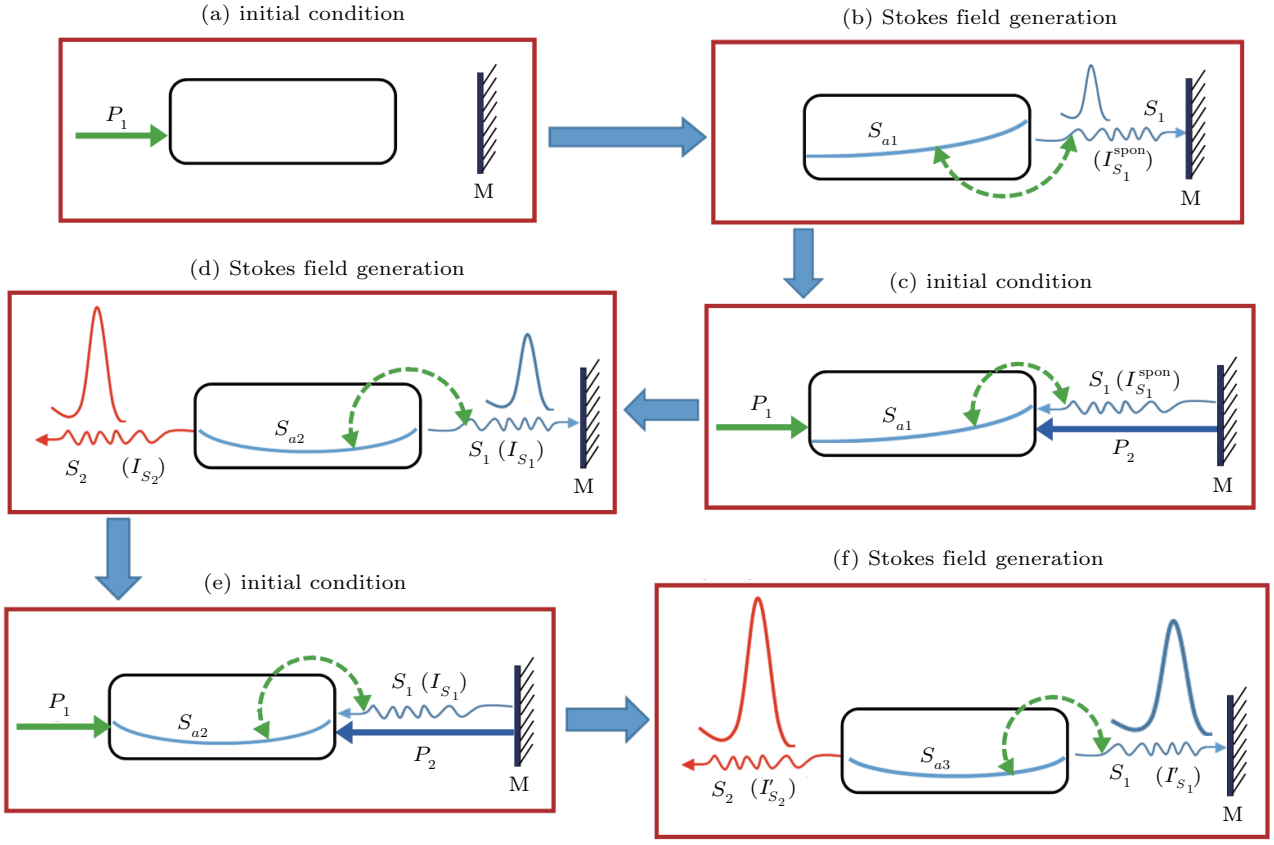


Fig. 2. (color online) The schematic diagram of cascade correlation-enhanced Raman scattering. The output intensities of Stokes fields are dependent on the initial conditions. (a) Experimental setup. The experimental setup is simple and the same with usual spontaneous Raman scattering in additional to a flat mirror. (b) The pump field P_1 leads to spontaneous emission of the Stokes field S_1, accompanied with the generation of atomic spin wave S_{a1}. (c) The atomic spin wave S_{a1} stays in the cell, the S_1 and P_1 fields propagate out together and both are reflected back to the atomic medium by a flat mirror M. (d) Two correlation-enhanced Raman scattering Stokes fields S_1 and S_2. Panels (a)–(d) form the first cycle and panels (e)–(f) form the second cycle. The cycles are repeated constantly in a period, and the intensity of Stokes field S_2 is coherent enhancement until it is saturated. The dashed curves (green) denote the correlation between the Stokes field and atomic spin wave.

The second to the fourth terms in Eq. (31) show the additional intensities generated by the initial seed light, prepared spin wave, and their correlation, which has been studied by us.[21] At the same time, the initial condition for Stokes field S_1 is changed and given by

$$\langle\hat{\mathscr{E}}^{\dagger}_{S_1}(0,t')\hat{\mathscr{E}}_{S_1}(0,t')\rangle = 0; \quad \langle\tilde{\mathscr{E}}^{\dagger}_{S_2}(s_L,t')\hat{\mathscr{E}}_{S_2}(s_L,t')\rangle \neq 0;$$
$$\langle\hat{S}^{\dagger}_a(z,0)\hat{S}_a(z,0)\rangle \neq 0, \quad \langle\tilde{\mathscr{E}}^{\dagger}_{S_2}(s,0)\tilde{\mathscr{E}}_{S_2}(s,0)\rangle = 0; \tag{36}$$

$$\langle \tilde{\mathscr{E}}^{\dagger}_{S_1}(0,t')\hat{S}^{\dagger}_a(s,0)\rangle \neq 0 \tag{37}$$

and the intensity of Stokes field S_1 is given by

$$I_{S_1} = I^{\rm spon}_{S_1} + I^{\rm add}_{S_1}, \tag{38}$$

where

$$\begin{aligned} I^{\rm add}_{S_1} = {} & \frac{|\chi_1\chi_2|^2}{4c^2|\Lambda|^2}\Big\{(\nu^*_+ \mathrm{e}^{-\lambda^*_+ t'} - \nu^*_- \mathrm{e}^{-\lambda^*_- t'}) \\ & \times (\nu_+ \mathrm{e}^{-\lambda_+ t'} - \nu_- \mathrm{e}^{-\lambda_- t'})\langle \hat{S}^{\dagger}_a(s,0)\hat{S}_a(s,0)\rangle \\ & + \frac{c^2}{4}|\nu_+\nu_-|^2 \int_0^{t'}\int_0^{t'} \mathrm{d}t''\mathrm{d}t'''(\mathrm{e}^{-\lambda^*_+(t'-t'')} \\ & - \mathrm{e}^{-\lambda^*_-(t'-t'')})(\mathrm{e}^{-\lambda_+(t'-t''')} - \mathrm{e}^{-\lambda_-(t'-t''')}) \\ & \times \langle \tilde{\mathscr{E}}^{\dagger}_{S_2}(s_L,t'')\tilde{\mathscr{E}}_{S_2}(s_L,t''')\rangle + \frac{c}{2}\int_0^{t'} \mathrm{d}t'' \\ & \times \big[\nu_+\nu_-(\mathrm{e}^{-\lambda_+(t'-t'')} - \mathrm{e}^{-\lambda_-(t'-t'')})(\nu^*_+ \mathrm{e}^{-\lambda^*_+ t'} \\ & - \nu^*_- \mathrm{e}^{-\lambda^*_- t'})\langle \hat{S}^{\dagger}_a(s,0)\tilde{\mathscr{E}}_{S_2}(s_L,t'')\rangle + \mathrm{H.c.}\big]\Big\}, \end{aligned} \tag{39}$$

where the mechanism of $I^{\rm add}_{S_1}$ is CERS and is the same as that of Stokes S_2. Figures 2(a)–2(d) form the first cycle. Different from the CERS[21], in Fig. 2(d) the correlated initial condition simultaneously leads to enhanced Stokes S_1 and S_2 generation, which is also obtained from Eq. (16). In the subsequent cycle shown in Figs. 2(e)–2(f), the intensity of Stokes field S_1 and S_2 are coherent enhancement due to the stronger initial conditions. After the cycle is repeated, the intensity of Stokes field S_2 increases to saturation. Therefore, we can obtain high efficient Raman conversion.[20]

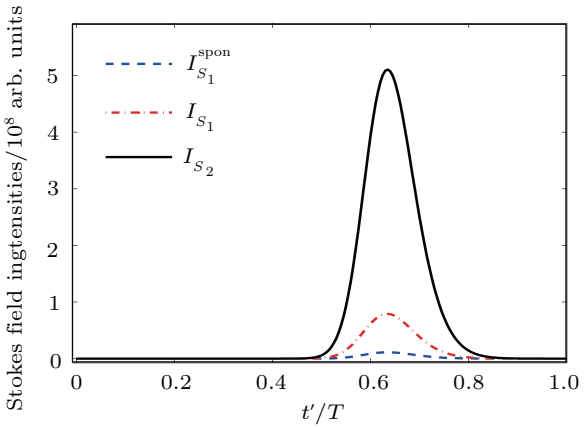


Fig. 3. (color online) The intensities of Stokes fields versus the dimensionless time in the first cycle. The parameters are as follows: $\Delta = 1.2$ GHz, $\gamma_{21} = 10$ kHz, $\gamma_{31} = 2\pi \times 5.746$ MHz, $Ng_1^2|\Omega_{P_{10}}|^2 TL/(c\Delta^2) = 8.5$, $\Omega_{P_{10}} = \Omega_{P_{20}}$, $T_1 = T_2 = T$.

Next, we will numerically calculate the intensities of $I^{\rm spon}_{S_1}$, I_{S_1}, and I_{S_2} using Gaussian-shaped write fields. Assume the write fields Ω_{P_1} and Ω_{P_2} have the following Gaussian shapes,

$$\Omega_{P_1}(t') = \Omega_{P_{10}}[\mathrm{e}^{-30(t'/T_1-0.5)^2} - \mathrm{e}^{-7.5}], \tag{40}$$

$$\Omega_{P_2}(t') = \Omega_{P_{20}}[\mathrm{e}^{-30(t'/T_2-0.5)^2} - \mathrm{e}^{-7.5}], \tag{41}$$

where T_1 and T_2 are the pulse durations of pump fields P_1 and P_2, respectively, and $\Omega_{P_{10}}$ and $\Omega_{P_{20}}$ are the coefficients. The intensities of Stokes fields as a function of the dimensionless time is shown in Fig. 3 in the first cycle. Due to correlation enhancement, the intensities of I_{S_1} and I_{S_2} are larger than the intensity $I^{\rm spon}_{S_1}$. The intensity of I_{S_2} is much larger than I_{S_1}, because the spatial distribution of the flipped atoms generated by the spontaneous process leads to that the intensity of counter-propagation case I_{S_2} is larger than that of co-propagation case I_{S_1}.[15]

4. Conclusion

In conclusion, we have given a theoretical model, termed as the cascade CERS, to describe the efficient Raman process. In the first cycle it is correlation-enhanced Raman scattering, i.e., injecting a seeded light field which is correlated with the initially prepared atomic spin excitation and driven by the quasi-standing-wave pump fields. The cycles are repeated constantly in a period, and the intensities of Stokes fields are coherent enhancement, then high efficient Raman conversion is achieved. Such a cascade correlation-induced enhanced Raman scattering process may find applications in a diversity of technological areas such as optical detection, metrology, imaging, precision spectroscopy, and so on.

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