

# STOKES–ANTI-STOKES ENTANGLEMENT IN STIMULATED RAMAN SCATTERING

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A cavity model of Raman scattering from phonons by an undepleted laser field is considered. The fields in the interaction are coupled to the boson reservoir that produces the damping mechanism in the model. An analysis for the origination of quantum entanglement between the Stokes and anti-Stokes fields, depending on their coupling constants with the reservoir bosons and on the state of the reservoir, is performed under the assumption of the initial coherent state of phonons.

Рассматривается резонаторная модель комбинационного рассеяния на фононах с классической лазерной накачкой. Взаимодействующие поля связаны с бозонным резервуаром, что обеспечивает механизм потерь в модели. Анализируются условия возникновения квантового перепутывания между стоксовым и антистоксовым полями в зависимости от их констант взаимодействия с бозонами в резервуаре и от состояния резервуара в предположении, что фононы изначально находились в когерентном состоянии.

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## INTRODUCTION

Stimulated Raman scattering is a useful method for spectroscopy in condensed matter physics and in examining fundamental aspects of quantum electrodynamics. Investigation of the nonclassical behavior of stimulated Raman scattering has been the subject of a number of papers [1, 2]. Recently, in view of the development of quantum information there has been arisen some interest in the study of the entanglement phenomenon between the fields in Raman scattering and estimation of the measure of their entanglement [3, 4].

In this paper, the entanglement initiation between scattered fields in stimulated Raman scattering with damping is considered with taking into account the dynamics of phonons. It allows one to study the quantum correlations in the model on conditions that phonons can initially be in a coherent state rather than in a chaotic one.

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## 1. MODEL

In this model, we assume an undepleted laser field of frequency  $\omega_L$ , and phase  $\phi_L$ , which can be treated classically. The fields in the interaction,  $\hat{a}_j$ , are the Stokes field, subscript  $S$ , anti-Stokes field, subscript  $A$ , and vibrational phonon field, subscript  $V$ ; their respective frequencies are  $\omega_S$ ,  $\omega_A$ , and  $\omega_V$ . The boson reservoir with multiple modes,  $\hat{b}_l^{(j)}$ , and frequencies  $\psi_l^{(j)}$ , provides a damping mechanism for the single-mode fields through its interaction with coupling constants  $\kappa_{jl}$ . The Hamiltonian of the system is given by [5]

$$\begin{aligned} \hat{H} = & \hbar\omega_V \hat{a}_V^\dagger \hat{a}_V + \hbar\omega_S \hat{a}_S^\dagger \hat{a}_S + \hbar\omega_A \hat{a}_A^\dagger \hat{a}_A + \hbar \sum_j \sum_l (\psi_l^{(j)} \hat{b}_l^{(j)\dagger} \hat{b}_l^{(j)} + \kappa_{jl} \hat{a}_j^\dagger \hat{b}_l^{(j)} + \kappa_{jl}^* \hat{a}_j \hat{b}_l^{(j)\dagger}) - \\ & (\hbar g_S \hat{a}_S^\dagger \hat{a}_V^\dagger e^{-i(\omega_L t + \phi_L)} + \hbar g_A \hat{a}_V \hat{a}_A^\dagger e^{-i(\omega_L t + \phi_L)} + \text{h.c.}), \quad j = V, S, A, \end{aligned} \quad (1)$$

where the laser–Stokes and laser–anti-Stokes coupling constants are  $g_S$  and  $g_A$ , respectively. The operators  $\hat{a}_j$  and  $\hat{b}_l^{(j)}$  satisfy the commutation relations

$$[\hat{a}_j, \hat{a}_k^\dagger] = \delta_{jk}, \quad [\hat{b}_l^{(j)}, \hat{b}_m^{(k)\dagger}] = \delta_{lm} \delta_{jk}, \quad j, k = V, S, A. \quad (2)$$

The frequencies  $\omega_L$ ,  $\omega_S$ ,  $\omega_A$ , and  $\omega_V$  are assumed to satisfy the resonance condition

$$\omega_S = \omega_L - \omega_V, \quad \omega_A = \omega_L + \omega_V. \quad (3)$$

The reservoir frequencies  $\psi_l^{(j)}$  are considered to be strongly coupled only to those radiation modes for which  $\psi_l^{(j)} \simeq \omega_j$ .

The Heisenberg equations of motion are given by

$$\begin{aligned} \frac{d\hat{a}_V}{dt} &= -i\omega_V \hat{a}_V + ig_S \hat{a}_S^\dagger e^{-i(\omega_L t - \phi_L)} + ig_A^* \hat{a}_A e^{i(\omega_L t - \phi_L)} - i \sum_l \kappa_{Vl} \hat{b}_l^{(V)}, \\ \frac{d\hat{a}_S}{dt} &= -i\omega_S \hat{a}_S + ig_S \hat{a}_V^\dagger e^{-i(\omega_L t - \phi_L)} - i \sum_l \kappa_{Sl} \hat{b}_l^{(S)}, \\ \frac{d\hat{a}_A}{dt} &= -i\omega_A \hat{a}_A + ig_A \hat{a}_V e^{-i(\omega_L t - \phi_L)} - i \sum_l \kappa_{Al} \hat{b}_l^{(A)}, \\ \frac{d\hat{b}_l^{(j)}}{dt} &= -i\psi_l^{(j)} \hat{b}_l^{(j)} - i\kappa_{Vl}^* \hat{a}_j. \end{aligned} \quad (4)$$

In the Markovian approximation, the equations are simplified by eliminating the  $\hat{b}_l^{(j)}$  operators. These equations in the interaction picture  $\hat{a}_j(t) = \hat{A}_j(t) e^{-i\omega_j t}$  are reduced into

$$\begin{aligned} \frac{d\hat{A}_V}{dt} &= -\frac{1}{2}\gamma_V \hat{A}_V + ig_S e^{i\psi_L} \hat{A}_S^\dagger + ig_A^* e^{-i\psi_L} \hat{A}_A + \hat{F}_A, \\ \frac{d\hat{A}_S}{dt} &= -\frac{1}{2}\gamma_S \hat{A}_S + ig_S e^{-i\psi_L} \hat{A}_V^\dagger + \hat{F}_S, \\ \frac{d\hat{A}_A}{dt} &= -\frac{1}{2}\gamma_A \hat{A}_A + ig_A e^{i\psi_L} \hat{A}_V + \hat{F}_A, \end{aligned} \quad (5)$$

where the Langevin forces due to the boson fields are

$$\begin{aligned}
\hat{F}_V &= i \sum_l \kappa_{Vl}^* \hat{b}_l^{(V)}(0) e^{-i(\psi_l^{(V)} - \omega_V)t}, \\
\hat{F}_S &= i \sum_l \kappa_{Sl} \hat{b}_l^{(S)}(0) e^{-i(\psi_l^{(S)} - \omega_S)t}, \\
\hat{F}_A &= i \sum_l \kappa_{Al}^* \hat{b}_l^{(A)}(0) e^{-i(\psi_l^{(A)} - \omega_A)t},
\end{aligned} \tag{6}$$

which satisfy the quantum fluctuation–dissipation theorem, and  $\gamma_j = 2\pi|\kappa_j(\omega_j)|^2\rho(\omega_j)$  are the damping constants,  $\rho(\omega_j)$  is the density function of the damping oscillators.

The equations of motion in Eqs.(5) can be directly solved using the Laplace transform method and the solutions are given as

$$\begin{aligned}
\hat{A}_V(t) &= \lambda_V(t)\hat{a}_V + \lambda_S(t)\hat{a}_S^\dagger + \lambda_A(t)\hat{a}_A + \sum_l [\Lambda_{Vl}(t)\hat{b}_l^{(V)} + \Lambda_{Sl}(t)\hat{b}_l^{(S)\dagger} + \Lambda_{Al}(t)\hat{b}_l^{(A)}], \\
\hat{A}_S(t) &= \mu_V(t)\hat{a}_V^\dagger + \mu_S(t)\hat{a}_S + \mu_A(t)\hat{a}_A^\dagger + \sum_l [M_{Vl}(t)\hat{b}_l^{(V)\dagger} + M_{Sl}(t)\hat{b}_l^{(S)} + M_{Al}(t)\hat{b}_l^{(A)\dagger}], \\
\hat{A}_A(t) &= \nu_V(t)\hat{a}_V + \nu_S(t)\hat{a}_S^\dagger + \nu_A(t)\hat{a}_A + \sum_l [N_{Vl}(t)\hat{b}_l^{(V)} + N_{Sl}(t)\hat{b}_l^{(S)\dagger} + N_{Al}(t)\hat{b}_l^{(A)}],
\end{aligned}$$

where the operators on the right-hand side are with respect to the initial state and explicit forms of the time-dependent functions are written down in [6].

## 2. THE WIGNER FUNCTION

The dynamics of the fields can be described by means of the Wigner function. Let us define the symmetric characteristic function for the Stokes, anti-Stokes and phonon fields with the assumption that they are initially in coherent states  $|\alpha_S\rangle$ ,  $|\alpha_A\rangle$ , and  $|\alpha_V\rangle$ , respectively, whereas the reservoir is found in a chaotic state with the boson mean number  $\langle n \rangle$ :

$$\begin{aligned}
\chi_{\text{sym}}(\beta_V, \beta_S, \beta_A; t) &= \text{Tr} \left\{ \hat{\rho}(0) \exp \left[ \beta_V \hat{A}_V^\dagger(t) + \beta_S \hat{A}_S^\dagger(t) + \beta_A \hat{A}_A^\dagger(t) - \text{h.c.} \right] \right\} = \\
&= \exp \left\{ -B_V(t)|\beta_V|^2 - B_S(t)|\beta_S|^2 - B_A(t)|\beta_A|^2 + \right. \\
&+ [D_{VS}^*(t)\beta_V\beta_S + D_{SA}^*(t)\beta_S\beta_A + D_{VA}(t)\beta_V\beta_A^* + \text{c.c.}] + \\
&\quad \left. + [\alpha_V^*(t)\beta_S + \alpha_S^*(t)\beta_V + \alpha_A^*(t)\beta_A - \text{c.c.}] \right\}, \tag{7}
\end{aligned}$$

where

$$\begin{aligned}
\alpha_V(t) &= \lambda_V(t)\alpha_V + \lambda_S(t)\alpha_S^* + \lambda_A(t)\alpha_A, \\
\alpha_S(t) &= \mu_V(t)\alpha_V^* + \mu_S(t)\alpha_S + \mu_A(t)\alpha_A^*, \\
\alpha_A(t) &= \nu_V(t)\alpha_V + \nu_S(t)\alpha_S^* + \nu_A(t)\alpha_A,
\end{aligned} \tag{8}$$

and the functions at bilinear variables for  $\gamma_V = \gamma_S = \gamma_A = \gamma$  are

$$\begin{aligned}
 B_V(t) &= |\lambda_S(t)|^2 + \langle n \rangle (1 - e^{-\gamma t}) + (2\langle n \rangle + 1) \sum_l |\Lambda_{Sl}(t)|^2, \\
 B_S(t) &= |\mu_V(t)|^2 + |\mu_A(t)|^2 - (\langle n \rangle + 1)(1 - e^{-\gamma t}) \\
 &\quad + (2\langle n \rangle + 1) \sum_l |M_{Sl}(t)|^2, \\
 B_A(t) &= |\nu_S(t)|^2 + \langle n \rangle (1 - e^{-\gamma t}) + (2\langle n \rangle + 1) \sum_l |N_{Sl}(t)|^2, \\
 D_{VS}(t) &= \lambda_S(t) \bar{\mu}_S(t) + (2\langle n \rangle + 1) \sum_l M_{Sl}(t) \Lambda_{Sl}(t), \\
 D_{SA}(t) &= \mu_S(t) \nu_S(t) + (2\langle n \rangle + 1) \sum_l M_{Sl}(t) N_{Sl}(t), \\
 D_{VA}(t) &= -\nu_S^*(t) \lambda_S(t) + (2\langle n \rangle + 1) \sum_l N_{Sl}^*(t) \Lambda_{Sl}(t).
 \end{aligned} \tag{9}$$

Then the Wigner function of the fields under consideration reads as

$$\begin{aligned}
 W(\xi_V, \xi_S, \xi_A; t) &= \frac{1}{\pi^6} \int d^2 \beta_V d^2 \beta_S d^2 \beta_A \exp [(\xi_V \beta_V^* - \xi_V^* \beta_V)] + \\
 &\quad + (\xi_S \beta_S^* - \xi_S^* \beta_S) + (\xi_A \beta_A^* - \xi_A^* \beta_A)] \chi_{\text{sym}}(\beta_V, \beta_S, \beta_A; t). \tag{10}
 \end{aligned}$$

In order to describe the dynamics of the scattered Stokes and anti-Stokes fields, it is necessary to pass on to the marginal Wigner function as a result of integration of (10) over the phonon variable

$$\begin{aligned}
 W(\xi_S, \xi_A; t) &= \int d^2 \xi_V W(\xi_V, \xi_S, \xi_A; t) = \frac{1}{\pi^2 \mathcal{N}(t)} \exp \left\{ -\frac{1}{\mathcal{N}(t)} \left( B_A(t) |\xi_S - \alpha_S(t)|^2 + \right. \right. \\
 &\quad \left. \left. + B_S(t) |\xi_A - \alpha_A(t)|^2 - [D_{SA}^*(t) (\xi_S - \alpha_S(t)) (\xi_A - \alpha_S(t)) + \text{c.c.}] \right) \right\} \tag{11}
 \end{aligned}$$

with the normalization

$$\mathcal{N}(t) = B_S(t) B_A(t) - |D_{SA}(t)|^2. \tag{12}$$

### 3. STOKES–ANTI-STOKES ENTANGLEMENT

The Wigner function (11) of the Gaussian type can be represented in the form

$$W(\boldsymbol{\gamma}; t) = \frac{1}{4\pi^2 \sqrt{\det \mathbf{V}(t)}} \exp \left\{ -\frac{1}{2} \boldsymbol{\gamma}^T \mathbf{V}^{-1}(t) \boldsymbol{\gamma} \right\} \tag{13}$$

with the help of the covariance matrix

$$\mathbf{V} = \begin{pmatrix} \mathbf{X} & \mathbf{Z} \\ \mathbf{Z}^T & \mathbf{Y} \end{pmatrix} = \begin{pmatrix} B_A & 0 & \text{Re } D_{SA} & \text{Im } D_{SA} \\ 0 & B_A & \text{Im } D_{SA} & -\text{Re } D_{SA} \\ \text{Re } D_{SA} & \text{Im } D_{SA} & B_S & 0 \\ \text{Im } D_{SA} & -\text{Re } D_{SA} & 0 & B_S \end{pmatrix}, \tag{14}$$

and the vector  $\gamma^T = (q_S, p_S, q_A, p_A)$  is composed by the real elements  $q_i = \sqrt{2}\text{Re}[\xi_i - \alpha_i(t)]$  and  $p_i = \sqrt{2}\text{Im}[\xi_i - \alpha_i(t)]$ .

The covariance matrix (14) can be used to reveal the presence of entanglement in the Stokes–anti-Stokes subsystem during its evolution by taking into consideration the measure of entanglement for Gaussian states suggested in [7]. This measure is completely defined by the symplectic spectrum of the partial transform of the covariance matrix that for the form (14) leads to the following expression of the logarithmic negativity [4]:

$$E = -\frac{1}{2} \log_2 [4f(\mathbf{V})], \quad (15)$$

where

$$\begin{aligned} f(\mathbf{V}) &= \frac{\det \mathbf{X} + \det \mathbf{Y}}{2} - \det \mathbf{Z} - \sqrt{\left( \frac{\det \mathbf{X} + \det \mathbf{Y}}{2} - \det \mathbf{Z} \right)^2 - \det \mathbf{V}} = \\ &= \frac{1}{2} \left[ B_S^2 + B_A^2 + 2|D_{SA}|^2 - (B_S + B_A) \sqrt{(B_S - B_A)^2 + 4|D_{SA}|^2} \right]. \end{aligned} \quad (16)$$

It shows the measure of entanglement for  $E > 0$ , and the case of  $E \leq 0$  indicates the separability of the subsystem state.

The behavior of the logarithmic negativity (15) depending on the interaction time and the boson mean number in the reservoir for various damping constants is displayed in Fig. 1 for the case  $g_S < g_A$ . If the damping constant  $\gamma$  is much less than the field–pump constants  $g_{S,A}$ ,  $E$  shows slightly decaying oscillations with definite regions where it takes on positive values indicating the entanglement in the subsystem of the Stokes and anti-Stokes fields, Fig. 1, *a*. These regions are shrunk when the boson mean number increases that demonstrates the phenomenon of the entanglement destruction by the reservoir noise. A similar periodicity in time of the antibunching effect between the Stokes and anti-Stokes modes was observed in [5]. As the damping constant  $\gamma$  increases, an oscillatory character of  $E$  still remains but the rate of the oscillation damping of the logarithmic negativity turns out to be more noticeable, Fig. 1, *b*. In case the damping constant  $\gamma$  becomes comparable with the field–pump constants  $g_{S,A}$ , the logarithmic negativity no longer displays oscillations and keeps moderate positive values only in a short range of variation of the boson mean number near to zero, Fig. 1, *c*.

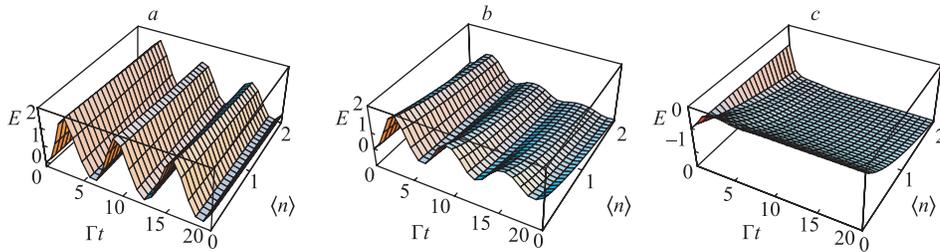


Fig. 1. Logarithmic negativity  $E$  (15) vs. the scaled time  $\Gamma t$  and boson mean number  $\langle n \rangle$  in the reservoir for  $g_A = 2g_S = 2 \cdot 10^7 \text{ s}^{-1}$  ( $\Gamma = (g_A^2 - g_S^2)^{-1/2} \approx 0.58 \cdot 10^{-7} \text{ s}$ ) and  $\gamma = 10^5 \text{ s}^{-1}$  (a),  $\gamma = 10^6 \text{ s}^{-1}$  (b),  $\gamma = 10^7 \text{ s}^{-1}$  (c)

In conclusion, it is worth noting that the peculiar pattern of entanglement between the Stokes and anti-Stokes fields found out in the paper may have promising applications in optical communication and quantum information processing.

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