# Phase quasi-integral for stimulated Raman scattering initiated by quantum fluctuations and statistics of solitonlike random pulses in a depleted pump

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A phase quasi-integral for stimulated Raman scattering (SRS) is found. The relative phase of laser, Stokes, and polarization waves is found to be nearly conserved during their propagation in Raman media. Due to this quasi-integral, the temporal statistics of solitonlike pulses in a depleted pump is defined by phase statistics in the linear stage of SRS.

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Stimulated Raman scattering (SRS) is a quantum macroscopic process starting from an unstable state. Any fluctuations, initial or boundary, classical or of quantum nature, lead to macroscopic output fluctuations of the Stokes amplitude. Manifestations of these fluctuations in the statistics of the Stokes pulse energy and its spectrum have been studied extensively for the linear stage of SRS (see review in  $[1]$ ).

Another fluctuation phenomenon arises at the nonlinear stage of SRS, i.e., the random spikes, the so-called solitonlike spikes, are observed in a depleted pump [2—4]. The nature of the spikes is attributed to the large phase flips of the relative phase of the pump, Stokes, and polarization waves [5]. This explanation is based on the results of Ref. [2], where spikes were obtained by deterministically induced phase flips into the Stokes seed and on the statement that such phase flips can arise due to the quantum noise [6]. On the other hand, it is expected that the sharp spikes in the pump-depleted region are connected with the well-known soliton solution of the SRS equation [7]. But the solution of the SRS equation is obtained in the case when the dephasing term is omitted from the polarization equation, whereas in all experimental realizations [2—4] the sharp spikes in the pump-depleted region have been observed in the case of essential dephasing. Besides, two questions arise: Why is the main role in sharp-spike initiation played by the phase flip, and not by the particular value of the relative phase? How is the phase switch connected with the possibility of sharpspike observation?

Here we show that the solitonlike spikes in the depleted region of the pump correspond to definite phase relations between Stokes, laser, and polarization waves for which the Stokes wave does not practically arise. The long-distance propagation of the spikes is due to the phase quasi-integral for SRS. The relative phase of laser, Stokes, and polarization waves is nearly conserved during propagation in Raman media. On the basis of this quasiintegral we calculate the temporal statistics of solitonlike spikes using the statistics of the phase at the linear stage of SRS.

# II. INTRODUCTION II. BASIC CONCEPTIONS

Stimulated Raman scattering initiated by' quantumnoise fluctuations can be described by different equations [5]. Here we will use the quantum equations corresponding to the normal ordering of the considered Stokes-field mean values [such as intensity of the Stokes field  $\langle E_S^-(z,t)E_S^+(z,t)\rangle$  [8]:

$$
\left[\frac{\partial}{\partial t} + c \frac{\partial}{\partial z}\right] E_S^+ = ig_z Q^- E_L^+ , \qquad (1)
$$

$$
\left[\frac{\partial}{\partial t} + c \frac{\partial}{\partial z}\right] E_L^+ = ig_z \left[\frac{\omega_L}{\omega_S}\right] Q^+ E_S^+ , \qquad (2)
$$

$$
\left(\frac{\partial}{\partial t} + \Gamma\right)Q^+ = ig_1 E_S^- E_L^+ + D^{1/2} F^+(z,t) ,\qquad (3)
$$

where  $g_z = 2\pi \omega_s Ng_1$ ,  $g_1$  is the susceptibility at the Stokes frequency,  $\omega_s$  and  $\omega_L$  are the frequencies of the Stokes and laser waves, respectively, N is the density of atoms in the ground state,  $T_2 = \Gamma^{-1}$  is the dephasing time,  $D = 2\Gamma / SN$  is the fluctuation intensity of the polarization due to the collision dephasing,  $S$  is the cross section of the excited medium volume, and  $F^+$  is the operator source of the fluctuations satisfying the relations

$$
\langle F^+(z,t)\rangle = 0 \tag{2a}
$$

$$
\langle F^+(z',t')F^-(z,t)\rangle = \delta(t-t')\delta(z-z') , \qquad (2b)
$$

$$
\langle F^+(z',t')F^+(z,t)\rangle = 0 \t , \t (2c)
$$

$$
F^{-}(z',t')F^{+}(z,t)\rangle = 0 , \qquad (2d)
$$

which correspond to the constant ground-state population.

Boundary and initial conditions can be formulated on he base of the physical situation, i.e., laser radiation being in a coherent state and having amplitude  $E_0(t - z/c)$ is incident on atomic medium stretching from  $z=0$  to  $z=L$ . Both the polarization and the radiation at laser frequency are in the vacuum state before the incident radiation arrival  $(t-z/c) = 0$ , i.e. [9],

$$
Q^+(z,t=0) = (D/2\Gamma)^{1/2}F_q^+(z) , \qquad (3a)
$$

$$
E_i^+(z, t=0) = (4\pi\omega_i/S)^{1/2} F_i^+(z) \quad (i = S, L) , \qquad (3b)
$$

where  $F_i^+(z)$  satisfy the equations similar to (2),  $F_i^+(z)$ with different  $i$  commutating.

The radiation at Stokes frequency is in the vacuum state at the boundary of the medium  $(z=0)$ 

$$
E_S^+(z=0,t)=(4\pi\omega_S/Sc)^{1/2}F_S^+(t) ,
$$
 (4a)

whereas the boundary condition for the laser radiation looks like

$$
E_L^+(z=0,t) = E_0(t) + (4\pi\omega_L / Sc)^{1/2} F_L^+(t)
$$
 (4b)

after the application of a displacement operator with the coherent amplitude  $E_0$  to Eq. (1).

Equation (1), along with conditions  $(2)$ – $(4)$ , constitutes a quantum problem of Raman scattering. Its comprehensive solution allowing for the commutation relations (2) is unknown. It is possible, however, to use approximate boundary conditions

$$
E_S^+(z=0,t) = E_i^+(z,t=0) = 0 \quad (i=S,L) ,
$$
 (5)

$$
E_L^+(z=0,t) = E_0(t) \tag{6}
$$



FIG. 1. Stochastic realization of the laser (a) and Stokes (b) intensity for the initial stage of SRS [just before going into saturated regime ( $gz = 35$ )], calculated using Eq. (1) for the case of compressed H<sub>2</sub> at  $p=50$  atm ( $\Gamma = 1.65 \times 10^8 p = 8.25 \times 10^9 s^{-1}$ .  $g_0 = 4\pi g_1 g_2/c^2 \Gamma = 2.5 \times 10^{-9}$  cm/W.  $S = 1$  mm<sup>2</sup>) and the Gaussian input laser field with the temporal width  $\tau_p = 30$  ns.

instead of (3b), (4a), and (4b), taking into account that due to (2c) the average of normal ordered products such as the intensity of the Stokes field  $\langle E_S^-(z,t)E_S^+(z,t) \rangle$ does not depend on the initial and boundary conditions  $E_{S,L}^{+}(z, t=0)$ ,  $E_{S}^{+}(z=0, t)$ , and  $F_{L}^{+}(t)$  (see [8] and [9]). Writing (6) we take into account that the number of laser photons  $n_{L\Delta V} = [ |E_L(t)|^2 / 4\pi \omega_L ]\Delta V$  in the minimal considered volume  $\Delta V = \Delta zS$  must be larger than unity  $[ |E_L(t)|^2 \gg 4\pi\omega_L / S\Delta z ]$ , or the elementary volume must be chosen larger than  $4\pi\omega_L |E_L(t)|^2$ . In this case, Eqs. (2b) and (2d) are used only, out of all Eqs. (2), and consequently, Eq. (1) is regarded as c numerical.

With such an approach, the system (1) can be solved numerically. Some of the realizations of laser  $\langle E_L^-(z,t)E_L^+(z,t)\rangle$  and Stokes  $[\langle E_S^-(z,t)E_S^+(z,t)\rangle]$  intensities corresponding to the different propagation distance z are presented in Figs.  $1-3$ . These realizations are

obtained for the Gaussian input laser pulse  
\n
$$
|E_0(t)|^2 = |E_0|^2 \exp\{-4(\ln 2)[(t - t_{int})/\tau_p]^2\},
$$
\n(7)

where  $\tau_p$  is the pulse time width, and for modeling of the stochastic terms  $F_i^+(z,t) = F_{ix}(z,t) + iF_{iy}(z,t)$ ,  $(i = s, q)$ stochastic terms  $F_i(x, t) = F_{ix}(z, t) + iF_{iy}(z, t)$ ,  $(i = s, q)$ <br>with the help of the random independent pairs of num-<br>bers ( $F_{ix}, F_{iy}$ ) obeying the Gaussian statistics.<br>For the initial state of the SBS when the laser is not

For the initial state of the SRS when the laser is not strongly depleted (gz  $\leq$  30, where  $g = 2g_1g_2|E_0|^2\Gamma_c$  is the



FIG. 2. Stochastic realization of the laser (a) and Stokes (b) intensity for the intermediate nonlinear stage of SRS ( $gz = 40$ ). Other parameters are the same as for Fig. 1.

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FIG. 3. Stochastic realization of the laser (a) and the Stokes (b) intensity for the strongly nonlinear stage of SRS ( $gz$  =45).<br>Other parameters are the same as for Fig. 1.<br> $\frac{\partial}{\partial \tau} + \Gamma \left| q^+ = ig_1 g_2 |E_0(\tau)|^2 S^+ \right|$ 

SRS gain coefficient, fluctuations produce rather smooth spikes in the Stokes intensity (Fig. 1).

For the intermediate (Fig. 2) and strongly nonlinear (Fig. 3) stages of SRS there are the solitonlike spikes in the depleted region of the laser intensity which correspond to the gaps (dark solitons) in the Stokes pulse. This correspondence reflects the conservations of the number of quanta,

$$
|E_L^+(z,t)|^2 + (\omega_L/\omega_S)|E_S^+(z,t)|^2 = |E_0(t-z/c)|^2.
$$
 (8)

As follows from the comparison of the spikes' maximum temporal position for the different propagation distances z, these values are approximately the same. The propagation distance of the solutionlike spikes is much larger than the length  $(cT_2)$ . It is for this reason that these spikes are called SRS solitons. So the main question concerning the physical nature of the SRS solitonlike spikes is, What is the reason for the conservation of the position of the spike maximum in the depleted region of the pump?

#### III. BLOCH-SPHERE REPRESENTATION

To answer this question it is convenient to introduce on the basis of Eq. (8) the variables

$$
E_L^+(z,t) = |E_0(t - z/c)| \cos(\Psi) \exp(i\phi_L) , \qquad (9a)
$$

$$
E_S^+(z,t) = (\omega_S/\omega_L)^{1/2} |E_0(t - z/c)| \sin(\Psi) \exp(i\phi_S),
$$
\n(9b)

and to make the well-known transformation [5,10]

$$
S^+ = X + iY = \sin(2\Psi)\exp[i(\phi_S - \phi_L)] \tag{10a}
$$

$$
S_z = Z = \cos^2 \Psi - \sin^2 \Psi \tag{10b}
$$

The value of  $S^+$  is proportional to the field amplitude product  $E_S^+E_L^-$ , whereas  $S_z$  is proportional to the difference of the laser and the Stokes quanta,<br> $E_L^+(z,t)|^2 - (\omega_L/\omega_S)|E_S^+(z,t)|^2$ . So the stochastic movement of the field variables can be represented on the sphere (Bloch sphere) as the vector  $(X, Y, Z)$  (Fig. 4). When Eqs. (10) are used in Eq. (1) and then in the moving frame  $(\tau = t - z/c)$  we obtain the Bloch-like equations

$$
ic\frac{\partial S^+}{\partial z} = q^+ S_z \t\t(11)
$$

$$
ic\frac{\partial S_z}{\partial z} = (q^-S^+ - q^+S^-)/2 \t\t(12)
$$

where  $S^+$  and  $S_z$  play the roles of the nondiagonal matrix element and the population inversion, respectively. These equations are supplemented with the stochastic equation for the Rabi frequency,  $q^+=2g_2(\omega_L/\omega_S)^{1/2}Q^+$ .

$$
\left[\frac{\partial}{\partial \tau} + \Gamma \right] q^+ = ig_1 g_2 |E_0(\tau)|^2 S^+ + 2g_2 (D \omega_L / \omega_S)^{1/2} F^+(z, \tau) .
$$
 (13)

The stochastic trajectories  $(X(\tau), Y(\tau), Z(\tau))$  on the Bloch sphere for the different propagation distances z are presented in Figs. 5 and 6. The system starts from the



FIG. 4. Bloch-sphere representation of the SRS field variables. Vector OS is defined by azimuth  $\phi_L - \phi_S$  and polar 2 $\Psi$ angles [Eqs. (9) and (10)]. The north pole  $(2\Psi=0)$  corresponds to the initial unstable state, whereas the south pole ( $2\Psi = \pi$ ) corresponds to the fully inverted stable state where there are no photons at laser frequency. The SRS field variables can also be presented as stereographical projection of the point S on the sphere onto the plane  $P$  (point  $R$ ).

unstable point ( $\Psi$ =0, the north pole on the Bloch sphere) and then moves along the trajectory and goes back into the unstable point. The movement near the south pole  $(2\Psi = \pi)$  corresponds to the laser depletion, whereas back and forth crossing of the equator corresponds to the spikes on Figs. 2 and 3. Note that the brokenness of the trajectories in some points is connected with the large scale of discretization for these points in the numerical simulation of Eqs. (1).

At first sight the stochastic trajectories in Figs. 5 and 6 corresponding to different propagation distances z are different. But if we look at the stereographical projections of these trajectories (Figs. 7 and 8), we find that these projections are almost similar to each other. The trajectories on the stereographical plane start from infinity, which corresponds to the north pole on the Bloch sphere. The main difference between the presented realizations is that most of the trajectory for the strongly nonlinear regime of the SRS (Fig. 8) lies inside the equator projection, that is, the reflection of the laser field depletion for this part.

# IV. RELATIVE PHASE CONSERVATION: TWO TYPES OF PHASE RELATIONS

The demonstrated similarity of the phase trajectories on the stereographical plane for different propagation distances suggests that the phase difference  $\phi_S - \phi_L$  is nearly conserved during propagation along the sample. To show this more clearly we present in Fig. 9 the equalphase  $\phi_S - \phi_L$  lines on the (z, t) plane. As follows from this figure the phase  $\phi_S - \phi_L = 0$  is nearly conserved along



FIG. 5. Stochastic trajectory  $X(t)$ ,  $Y(t)$ ,  $Z(t)$  on the Bloch sphere for the intermediate nonlinear stage of SRS ( $gz=40$ ). The system starts from the unstable point (north pole), then moves along the stochastic trajectory, and returns to the unstable point. Back and forth crossing of the equator corresponds to the spikes in temporal form of the Stokes and the laser pulses presented in Fig. 2.

 $\overline{Z}$  $\times$ 

FIG. 6. Stochastic trajectory  $X(t)$ ,  $Y(t)$ ,  $Z(t)$  on the Bloch sphere for the strongly nonlinear stage of SRS  $(gz=45)$ . The movement near the south pole corresponds to the laser depletion. Laser and Stokes intensities attributed to this stochastic trajectory are presented in Fig. 3.

the characteristics  $\tau = t - z/c = \text{const.}$  The largest deviations from these lines take place at the boundaries of the sample.

The quantitative explanation of the phase difference conservation can be done on the basis of Eqs.  $(11)$ – $(13)$ written in the form



FIG. 7. Stereographical projection of the trajectory presented in Fig. 5 ( $gz = 40$ ). The trajectory starts from infinity, which corresponds to the north pole on the Bloch sphere. Dashed circles are the stereographical projection of the equator of the sphere.

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 $\frac{\partial}{\partial \tau}$ 



FIG. 8. Stereographical projection of the trajectory presented in Fig. 6 ( $gz = 45$ ). The main difference between the trajectory realization in Fig. 7 and this trajectory is that the main part of the trajectory for the strongly nonlinear regime of SRS lies inside the equator projection (dashed line), which is the reflection of the laser field depletion for this part.



FIG. 9. Equal phase  $\phi_L - \phi_S$  lines on the (z,t) plane. The lines correspond to the zero value of the difference  $\phi_L - \phi_S$ . then according to Eqs. (14c) and (14d) in the vicinity of

$$
c\frac{\partial(\phi_S - \phi_L)}{\partial z} = |q^+ / S^+|S_z \cos(\phi_S - \phi_L + \phi_q), \qquad (14a)
$$

$$
\frac{\partial \phi_q}{\partial \tau} = g_1 g_2 |E_0(\tau)|^2 |S^+ / q^+| \cos(\phi_S - \phi_L + \phi_q)
$$
  
+Im(f^+ / |q^+|), \t(14b)

$$
c \frac{\partial |S^+|}{\partial z} = |q^+| S_z \sin(\phi_S - \phi_L + \phi_q) , \qquad (14c)
$$

$$
+\Gamma \Big| |q^+| = g_1 g_2 |E_0(\tau)|^2 |S^+|
$$
  

$$
\times \sin(\phi_S - \phi_L + \phi_q) + \text{Re} f^+ , \qquad (14d)
$$

where  $f^+ = 2g_2 (D\omega_L / \omega_S)^{1/2} F^+(z,\tau) \exp(-i\phi_q)$  and  $q^+=|q^+|$ exp( $i\phi_q$ ). Note that in this case we suppose that  $g_1$  and  $g_2$  are real.

### A. High-gradient strips

As follows from Eq. (14a), if at a point  $(z_0, \tau_0)$  the relative phase

$$
\Delta \phi = \phi_S - \phi_L + \phi_q = \pi/2 + \pi n \quad (n = 0, \pm 1, \pm 2, \dots) \tag{15}
$$

the phase difference  $\phi_S - \phi_L$  will be conserved in the vicinity of the point along the line parallel to the z axis [along the characteristic passed through the point  $(z_0, \tau_0)$ ] up to the point where  $\Delta \phi$  is changed. Besides, one can find from Eq. (14b) that on the characteristic the time derivative  $\partial \phi_q / \partial \tau$  will approximately equal zero for the small fluctuating term  $\text{Im}(f^+/\vert q^+\vert)$ , and this is indeed so. As follows from Eqs. (14b) and (14d) (see also [10], where the fluctuations along the characteristics have been studied), the intensity of the fiuctuations for the strongly nonlinear regime of SRS is of the order of

$$
\sigma^2 = v/g = 2(g_2/c)^2 (D\omega_L / \Gamma \omega_S) / g \tag{16}
$$

As an example for the Raman scattering in H<sub>2</sub> at  $p = 50$ atm and the laser cross section  $S=1$  mm<sup>2</sup> we have the the vector is  $v=10^{-15}$  m<sup>-1</sup>. Therefore the line  $t-z/c = \tau_0$  is characterized by smoothness of temporal behavior of the polarization phase  $\phi_q$  and as a consequence the same temporal behavior must be observed for the phase difference  $\phi_S - \phi_L$  (see Fig. 10).

So one can speak not about the lines with  $\Delta\phi = \pi/2 + \pi n$  but about the *strips* with finite temporal width where  $\Delta\phi \approx \pi/2+\pi n$ . Due to Eqs. (14c) and (14d) which where  $\Delta\varphi \approx \pi/2 + \pi\pi$ . Due to Eqs. (14c) and (14d)<br>the largest value of the gain [if  $S_z \sin(\phi_S - \phi_L + \phi_q) > 0$ ] or of the attenuation [if  $S_z \sin(\phi_s - \phi_L + \phi_q) < 0$ ] must be observed for these strips. That is why we call these strips high-gradient strips (HGS's).

#### B. Small-gain strips

If at the point  $(z_1, \tau_1)$  the relative phase

$$
\Delta \phi = \phi_S - \phi_L + \phi_q = \pi n \quad (n = 0, \pm 1, 2, \dots) \tag{17}
$$



FIG. 10. Stochastic realizations of the relative phase  $\Delta \phi = \phi_S + \phi_a - \phi_L$  for intermediate (a) and strongly nonlinear (b) stages of SRS. The time domains belonging to HGS's where  $\Delta \phi \approx \pi + \pi n$ , are much wider than the time domains belonging to SGS's, where  $\Delta \phi \approx \pi n$ . The comparison of the relative phase realizations for these two different propagation distances  $[gz = 40 \text{ (a)}, 45 \text{ (b)}]$  clearly shows the quasiconservation of the relative phase during propagation in the media.

the point the Stokes and laser intensities as well as the polarization modulus are not changed. If in addition the value of the polarization is small at the point  $(z_1, \tau_1)$  that corresponds to the boundary condition Eq. (3a) and small value of the coefficient  $D$ , then according to Eq. (14a) the phase difference  $\phi_S - \phi_L$  will be conserved along the characteristic passed through the point  $(z_1, \tau_1)$ . As follows from Eq. (14b), the temporal derivative  $\partial \phi_a / \partial \tau$  can be rather large on the characteristic  $t - z/c = \tau_1$ . Therefore the temporal width of the strip [small-gain strip (SGS)] will be much smaller than the width of the HGS (see Fig. 10). It should be noted that when due to the

fluctuations the value of  $\Delta \phi$  differs from  $\pi n$ , the phase difference  $\phi_S - \phi_L$  conservation on the SGS can be explained by two reasons: (i) zero value of the right-hand side of Eq. (14a) due to  $S_z=0$  in the intermediate regime of the SRS and (ii) space constriction of a SGS between the two nearest HGS's.

As a result one can represent the whole  $(z, t)$  plane for the SRS as a number of the interchanged SGS's and HGS's parallel to the z-ct axis. It is evident from this representation that during the propagation in the media the intensity of the Stokes and laser components will be changed first in HGS's and then in SGS's. Therefore the intensity shape of the Stokes and laser pulses will have the form of interchanged maxima and minima. The dips in the Stokes intensity and the spikes in the depleted laser intensity correspond to the SGS's. The long-distance propagation of the spikes is due to the fact that Raman scattering is not excited for SGS's. It should be noted that the same structure of the  $(z, t)$  plane has been discussed [11] in connection with the phase waves in superfluorescence.

# V. STATISTICS OF SPIKES IN DEPLETED LASER INTENSITY

Using the predicted conservation of the relative phase  $\Delta\phi = \phi_S - \phi_L + \phi_q$  at pulse propagation, and the relationship of the spikes in the depleted laser intensity to SGS's we can find the statistics of the spikes in the strongly nonlinear regime on the basis of the phase statistics in the linear regime of SRS. As follows from previous considerations the SGS is characterized not only by a definite value of the relative phase  $(\Delta \phi = \phi_s - \phi_L + \phi_q = \pi n)$  but also by a rather fast change of the polarization phase  $\phi_q$ and the phase difference  $\phi_S - \phi_L$  in the time scale. Therefore we can estimate the probability of the spike observation using the probability of the phase  $\phi_S - \phi_L$  (or  $\phi_q$ ) changing on large values in the linear regime of the SRS.

Because the statistics of the operator  $S^+$  in the linear regime coincides with the statistics of the Stokes field  $E<sub>S</sub><sup>+</sup>$ , which is Gaussian, we can use well-known relations [12] for the phase statistics of the Gaussian stochastic variables. So the probability of the two-time phase difference

$$
\phi = [\phi_S(t + \Delta t) - \phi_L(t + \Delta t)] - [\phi_S(t) - \phi_L(t)]
$$

to be in the interval

$$
\pi \geq |\phi| > \phi_0
$$

can be represented in the simple form

$$
C(\phi_0, t, \Delta t) = 1 - \frac{1}{\pi} \left[ \phi_0 + \frac{g^{(1,1)}(t, t + \Delta t) \sin \phi_0}{[1 - g^{(1,1)}(t, t + \Delta t) (\cos \phi_0)^2]^{1/2}} \arccos[-g^{(1,1)}(t, t + \Delta t) \cos \phi_0] \right],
$$
\n(18)

where

$$
g^{(1,1)}(t,t+\Delta t) = \frac{\langle E_S^-(t)E_S^+(t+\Delta t)\rangle}{[\langle E_S^-(t)E_S^+(t)\rangle\langle E_S^-(t+\Delta t)E_S^+(t+t\Delta t)\rangle]^{1/2}}
$$
(19)

is the normalized Stokes-field correlation function of the first order.

Using Eq. (18) we estimate the mean waiting time for changing of the phase  $\phi_S - \phi_L$  on large value  $(\phi_0 > \pi/2)$ as a time  $(\Delta t)$ , needed for increasing the probability

$$
C(t,\Delta t,\pi/2) = [1 - g^{(1,1)}(t,t+\Delta t)]/2, \qquad (20)
$$

up to the level  $(1-e^{-1})/2$ . [Note that the probability  $C(t, \Delta t, \pi/2)$  changes from 0 at  $\Delta t = 0$  to  $\frac{1}{2}$  at  $\Delta t \rightarrow \infty$ .] According to the definition the mean waiting time  $(\Delta t)_e$ for a large change of the phase  $\phi_S - \phi_L$  coincides with the instantaneous correlation time of the Stokes field. For the initial stage of SRS we have obtained (see Appendix) the approximate expression for correlation function  $g^{(1,1)}(t,t+\Delta t)$  which takes into account the transient effects in the case when  $\Gamma \tau_p >> 1$ :

$$
g^{(1,1)}(t, t + \Delta t) = \exp\{-\left[\Delta t / (\Delta t)_e\right]^2\},
$$
 (21)

where

$$
(\Delta t)_e = \frac{2}{\Gamma} \frac{\overline{\kappa}(t)}{\{\kappa[(t+\overline{t})/2]\}^{1/2}}
$$
 (22)

and

$$
\bar{t} = t - \frac{\kappa(t)}{2\Gamma} \tag{23}
$$

Here  $\kappa(t)=2g_1g_2z|E_0(t)|^2/\Gamma c=gz|E_0(t)/\overline{E}_0|^2$  is the instantaneous gain coefficient and  $|E_0|^2 = \int_{-\infty}^{\infty} d\tau |E_0(t)|^2$ is the integrated intensity of the input laser field.

The frequency of the solitonlike spikes appearance in the depleted region of the pump at a given time  $t$  can be calculated as

$$
v_{\rm sp} = \frac{1}{2} (\Delta t)^{-1} P_d(t, \eta) , \qquad (24)
$$

where  $P_d(t, \eta)$  is the characteristic function equal to unity when the mean value of the pump is depleted up to the fixed adopted level

$$
\eta = \omega_L \langle E_S^-(z, t) E_S^+(z, t) \rangle / \omega_S \langle E_L^-(z, t) E_L^+(z, t) \rangle , \quad (25)
$$

e.g.,  $\eta > 0.5$ , otherwise  $P_d(t, \eta) = 0$ . In other words, the value of  $v_{\rm sn}$  is defined only for the region where the second threshold of SRS is achieved. Using the approximate expression for the Stokes intensity (see Appendix) we can roughly estimate the second threshold from the inequality [10]

$$
\frac{e^{\kappa(\overline{t})}}{\left[\pi\kappa(\overline{t})\left[1+\Gamma^{-1}\frac{d\kappa(\overline{t})}{d\overline{t}}\right]\right]^{1/2}} \geq 2\sigma^{-2}, \qquad (26)
$$

with

$$
\sigma^{-2} \equiv \frac{g}{\nu} \equiv (|\overline{E}_0|^2 / 4\pi \omega_L) \Delta V_\Gamma
$$

being the number of input laser photons in the volume  $\Delta V_{\Gamma} = Sc/2\Gamma$ . Note that the factor  $\frac{1}{2}$  in Eq. (24) is due to the same factor in Eq. (20) defining the probability  $C(t, \Delta t, \pi/2)$ .

On the basis of Eqs.  $(19)$ – $(23)$  we have calculated the



FIG. 11. Normalized time-dependent frequency  $2\Gamma$  of large phase  $\phi_s - \phi_l$  changes for the Gaussian form of the laser input (3) at different propagation distances  $gz = 25$  (1); 45 (2) with the correlation time  $(\Delta t)_e$  calculated from Eqs. (A2)-(A4). Dotted lines correspond to the approximate Eq. (22). Two vertical lines mark two moments  $(T_F$  and  $T_B$ ) when the second threshold of SRS is achieved;  $T_F$  and  $T_B$  have been found from Eq. (26) at  $\sigma^{-2} = 10^{16}$ .  $\tau_p$  is the laser input time width. The circles mark the reverse time differences  $(t_i - t_{i-1})$  between the nearest values  $t_i$  at which  $\Delta \phi = \pi n$ , obtained from numerical simulations of Eq. (1).

time-dependent frequency  $\frac{1}{2}(\Delta t)^{-1}$  of large phase changes for the Gaussian form of the laser input [Eq. (7)] for different propagation distances (Fig. 11). The region where  $P_d(t, \eta)$  is equal to unity is located between the two vertical lines. In this region the frequency of large phase changes will coincide with the frequency of the solitonlike spikes in the depleted laser pulse. It should be noted that the observed small asymmetry in the temporal dependence of  $\frac{1}{2}(\Delta t)^{-1}$  arises due to the fact that the correlation time of the Stokes wave and the instantaneous gain coefficient  $\kappa(t)$  became asymmetrical in the case of nonrectangular laser pulse. As follows from Eqs. (22) and (23), these values are slightly smaller in the first part of the symmetrical pulse.

The circles in Fig. 11 denote reverse time differences  $(t_i - t_{i-1})^{-1}$  between the nearest values  $t_i$  at which  $\Delta \phi = \pi n$ , obtained from numerical simulations of Eq. (1). The rather good agreement between this straightforward calculation of the frequency of large phase changes and simple formula (22) is an additional argument in favor of our main reasoning about the quasiconversation of relative phase at the pulses propagation during SRS.

## VI. SUMMARY

Using straightforward numerical calculations of stochastic SRS equations together with the Bloch-sphere representation for SRS, as well as stereographical projections of stochastic trajectories, we have shown that the relative phase of the laser, Stokes, and polarization waves is nearly conserved during their propagation in Raman media.

We have shown that two types of regions with different

phase relations are created: the strips stretching along the characteristics  $(t - z/h = const)$  with nearly zero relative phase  $\Delta \phi$  ( $\Delta \phi \approx \pi n$ ) and parallel strips with the  $\pi/2$ phase difference ( $\Delta \phi \approx \pi/2+\pi n$ ). The first type of strips are characterized by a small gain [small-gain strips (SGS's)], a large value of the time derivative of the polarization wave phase  $(d\phi_q / d\tau)$  as well as of the derivative of the laser and Stokes phase difference  $[d(\phi_S - \phi_L)/d\tau]$ , and by a small temporal width of the strips. The second type of strips have the largest (for possible phase relations) value of gradient for the Stokes and laser intensities [high-gradient strips (HGS's)], small time derivatives of the phases  $d\phi_q / d\tau$  and  $d(\phi_L - \phi_S) / d\tau$ , and a rather wide temporal width of the strips. The alternation of SGS's and HGS's in time will be detected as temporal spikes in the depleted laser pulse, the time positions of the spikes coinciding with the positions of the SGS's.

Using the known solution for SRS in the linear regime we have obtained simple formulae for the frequency of the solitonlike spikes in the depleted laser pulse intensity for the highly nonlinear regime of scattering. The obtained relation agrees with the numerical calculations of the starting equation, and with the preliminary experimental results [13] obtained for the solitonlike statistics at the SRS in  $H_2$  gas. It should be noted that the predicted phase conservation can be observed not only in the temporal but also in the spectral fluctuations of the Stokes light as measured in [14].

In conclusion I want to mention intriguing problems of statistical properties of the soliton details such as amplitude and temporal width and their connection with the relative phase change statistics. Theoretical and experimental investigations of the problems are now in progress.

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#### APPENDIX

In this appendix we consider the Stokes-field correlation function of the first order and the mean intensity of the Stokes field in the linear steady-state regime of SRS without appealing to the rectangular-pulse-form approximation.

#### 1. Two-time correlation function

It is well known from [1,8] that the temporal correlation function of the Stokes field in the linear regime  $E^{+}_{L}(t) = E_{0}(t)$  ] is

$$
G^{(1,1)}(z,t; z,t') = \langle E_S^-(z,t)E_S^+(z,t') \rangle
$$
  
=  $(\omega_S / 4\omega_L)E_0(t)E_0^*(t')$   
 $\times \langle S^-(z,t)S^+(z,t') \rangle$  (A1)

where

$$
\langle S^-(z,t)S^+(z,t')\rangle = vze^{-\Gamma(t+t')} \left[ G_z(t,t';0) + 2\Gamma \int_0^{t'} d\tau \, e^{2\Gamma \tau} G_z(t,t';\tau) \right] \quad (t > t') \tag{A2}
$$

with the Green function 
$$
G_z(t, t'; \tau)
$$
 defined by  
\n
$$
G_z(t, t'; \tau) = \frac{2}{\beta(t, t')} [\beta^{1/2}(t, \tau) I_1(\beta^{1/2}(t, \tau)) I_0(\beta^{1/2}(t', \tau)) - (t \leftrightarrow t')],
$$
\n(A3)

where  $I_n(x)$  is the modified Bessel function and

$$
\beta(t,t') = 4z \frac{g_1 g_2}{c} \int_{t'}^{t} |E_0(\tau)|^2 d\tau .
$$
 (A4)

The value of  $vz/4$  in (A2) with v given by Eq. (16) is equal to the spontaneous Raman scattering transformation coefficient

$$
\eta_0 = [\omega_L \langle |E_S^+|^2 \rangle / \omega_S |E_0(\tau)|^2]_{E_0 \to 0} = vz / 4. \quad (A5)
$$

Since we consider the times  $t, t' \gg 1/\Gamma$ , then the main contribution to the correlation function  $G^{(1,1)}$  will be made by the second term in Eq. (A2). Besides, we use the fact that the contribution to the second term will be made by the temporal region where  $\beta(t, \tau)$  and  $\beta(t', \tau)$  are much larger than unity. So, on the basis of these assumptions and utilization of the asymptotic expansion of the Bessel functions  $I(x)$ , we find

$$
\langle S^-(z,t)S^+(z,t')\rangle \approx \frac{\nu z}{\pi} e^{-\Gamma(t-t')} 2\Gamma \int_0^{t'} d\tau e^{-2\Gamma(t'-\tau)+\beta^{1/2}(t,\tau)+\beta^{1/2}(t',\tau)}/\Delta(t,t',\tau)
$$
(A6)

where

$$
\Delta(t,t',\tau) = [\beta^{1/2}(t,\tau) + \beta^{1/2}(t',\tau)]\beta^{1/4}(t,\tau)\beta^{1/4}(t',\tau) .
$$

The integral (A6) can be evaluated in the case of high gain,

where  $|\overline{E}_0|^2 = \int_{-\infty}^{\infty} d\tau |E_0(\tau)|^2$  is the integrated intensity of the input laser field. Using the Laplace asymptotic method of integration  $[15]$  we obtain

$$
g^{(1,1)}(t, t + \Delta t) = \exp\{-[\Delta t/(\Delta t)_e]^2\}, \qquad (A7)
$$

 $g_Z = \frac{2g_1g_2}{\Gamma c} |\bar{E}_0|^2 z$ ,

where

$$
\left(\Delta t\right)_e = \frac{1}{\Gamma} \frac{\kappa(\overline{t})}{\left[\kappa((t+\overline{t})/2)\right]^{1/2}} \,, \tag{A8}
$$

and [see also  $(A14)$  and  $(A15)$ ]

$$
\overline{t} = t - \frac{\kappa(t)}{2\Gamma} \tag{A9}
$$

where  $\kappa(t) = 2g_1g_2|E_0(t)|^2/\Gamma c \equiv gz|E_0(t)/\overline{E}_0|^2$  is the in-<br>stantaneous gain coefficient  $\left[2\Gamma \int_{\overline{t}}^t \kappa(t')dt'\right]$ 

# 2. Mean intensity of the Stokes field (Second SRS threshold)

The mean intensity of the Stokes field is given by Eqs.  $(A1)$ – $(A4)$  at  $t = t'$ , with

$$
G_z(t,t;\tau) = I_0^2(\beta^{1/2}(t,\tau)) - I_1^2(\beta^{1/2}(t,\tau)) . \tag{A10}
$$

In the case when  $\Gamma t >> 1$  the laser-to-Stokes transformation coefficient (25) is defined by

$$
\eta = \frac{1}{4} \langle S^-(z, t) S^+(z, t) \rangle
$$
\n
$$
\equiv \frac{1}{4} \langle \sin^2 2\Psi \rangle
$$
\n
$$
= \frac{1}{4} \nu z 2 \Gamma \int_0^t d\tau e^{-2\Gamma(t-\tau)} G_z(t, t; \tau) .
$$
\n(A11) equal volume

The latter integral can be evaluated approximately to give in the high-gain limit  $(gz \gg 1)$ 

$$
\eta = \frac{\nu}{4g} \frac{e^{\kappa(\bar{t})}}{\{\pi \kappa(\bar{t})[1 + \Gamma^{-1} d\kappa(\bar{t})/d\bar{t}]\}^{1/2}} , \qquad (A12)
$$

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where

$$
\kappa(\overline{t}) = gz |E_0(\overline{t})/\overline{E}_0|^2
$$
\n(A13)

is the instantaneous SRS increment and the time  $\bar{t}$  is given by the equation

$$
-\frac{\kappa(\bar{t})}{\left[2\Gamma\int_{\bar{t}}^t \kappa(t')dt'\right]^{1/2}}=0.
$$
 (A14)

The approximate solution of (A14) is

$$
\overline{t} = t - \frac{\kappa(t)}{2\Gamma} \tag{A15}
$$

The second SRS threshold [10] is achieved when the laser-to-Stokes transformation coefficient  $\eta$  becomes aser-to-stokes transformation coefficient  $\eta$  occomes arger than  $\frac{1}{2}$ . In this case the linear approximation cannot be used to obtain the exact relations. But taking into account the relatively large value of the parameter

$$
\sigma^{-2} \equiv \frac{g}{\nu} \equiv (|\overline{E}_0|^2 / 4\pi \omega_L) \Delta V_\Gamma
$$
 (A16)

equal to the number of the input laser photons in the volume  $\Delta V_{\Gamma}$  = Sc/2 $\Gamma$ , we obtain that the linear approximation gives rather good estimation for the second SRS threshold,

$$
\frac{e^{\kappa(\bar{t})}}{\{\pi\kappa(\bar{t})[1+\Gamma^{-1}d\kappa(\bar{t})/d\bar{t}]\}^{1/2}} \geq 2\sigma^{-2} .
$$
 (A17)

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