Inverse-Scattering Transform of Stimulated Raman and Brillouin Scattering in the Quasi-Steady-State Regime

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Stimulated Raman and Brillouin scattering in the quasi-steady-state (QSS) regime are analyzed by the inverse-scattering transform (IST) with nonoverlapping square wave packets and second-order Zakharov-Shabat scattering problem. It is shown that the important QSS characteristics, namely, the number of pulses required for establishing the QSS condition, the Stokes growth, and the development of polarization are readily calculated using IST. These results are usually obtained by extensive numerical computations which, however, do not lead to their fundamental determinations. The IST results are found to be in excellent agreement with experimental observations and numerical computations.

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Stimulated Raman (SRS) and Brillouin (SBS) scattering with high conversion efficiencies are frequently used for amplifier-target decoupling,[1] compression of pulses along the time axis, and energy extraction from poor-quality long pumps into high-spatial-quality short extraction beams.[2–6] In addition, the phase-conjugation properties of SRS-SBS mirrors are extensively used for correcting wave-front distortions.[7,8] Large-reflectivity SRS-SBS experiments typically employ 10-ns or wider pumps. However, there are important applications where narrower pumps are required, for example, in laser fusion where high-power Nd-doped yttrium-aluminum-garnet (Nd:YAlG) (1.06 μm) or iodine (1.32 μm) systems are used with 1-ns or narrower pumps. This results in highly transient scattering with detrimental effects on gain, reflectivity, and threshold power. For short pulses, the geometric conditions for large reflection coefficients and good wave-front reversal properties become mutually exclusive[9] unless the pump is focused hard, in which case additional complications arise such as competition with undesired nonlinear processes, self-focusing, and the possibility of breakdown of the optical medium at the lens focus. Thus, narrow pumps have severe consequences for amplifier isolation, efficient high-power amplification, and pulse compression. This is particularly true when highly compressed gases are used since the gas breakdown threshold decreases with increasing pressure.

For these reasons, the recent trend is toward quasi-steady-state scattering[10–12] (QSSS) which uses a repetitive periodic sequence of pulses as the pump. The idea was first mentioned by Carman, Shimizu, Wang, and Bloembergen.[13] In QSSS, the gain increases with each pulse, leading to an equilibrium condition of gain saturation. Thus QSSS enhances gain in highly transient scattering processes and provides a means for keeping the threshold below that of the medium breakdown.

A proper analysis of SRS and SBS in the QSS regime requires that the coupled set of three-wave resonant-interaction (3WRI) equations corresponding to these processes be solved. However, because of the quadratic nonlinearity in the light fields, such a solution is difficult and various numerical procedures are, therefore, resorted to. Although important in understanding the characteristics of QSSS, numerical procedures do not reveal the fundamental nature of the solutions. Thus important QSSS considerations such as the number of pulses required to attain the QSS condition, the Stokes growth, and the medium polarization before this regime is established cannot be predicted by direct integration.[11]

It is demonstrated in this Letter that the inverse-scattering transform,[14–16] (IST) which solves a 3WRI system exactly, allows such a fundamental determination of the QSS characteristics of SRS and SBS processes which would otherwise have to be obtained by elaborate numerical integration. It is shown that a simple iterative application of IST yields the Stokes growth for each successive pulse in the QSS pump pulse train, the number of pulses required to establish the QSS regime, and the time development of the medium polarization. Finally, the simple and readily obtainable results of IST are compared to recent experimental observations and numerical studies.[11,12]

For IST analysis, it is convenient to write the SRS-SBS equations in the Benney-Newell[17] canonical form

$$\Box_j Q_j = i\gamma_j \prod_1^3 Q_k^*, \quad j,k=1,2,3, \quad k\neq j. \tag{1}$$

where

$$Q_1 = i[\kappa_\xi \kappa_l]^{1/2} E_s^*, \quad Q_2 = i[\kappa_s \kappa_l]^{1/2} \xi^*,$$

$$Q_3 = -i[\kappa_\xi \kappa_s]^{1/2} E_l,$$

$(\gamma_1,\gamma_2,\gamma_3)=(-1,-1,+1)$, and the asterisk denotes complex conjugation. Subscripts 1 (s), 2, and 3 (l) refer to SBS, material excitation, and pump envelope, respectively. $\square=\partial/\partial t+v(\partial/\partial x)$ is the wave operator in the slowly-varying-envelope approximation. $v_1(=-v_3)<v_2<v_3$, $\xi_{\rm SRS}=i\zeta$, ζ is the amplitude of the expectation value of the displacement operator of a normal vibrational coordinate, $\kappa_\xi=(1/4m\omega_R)\partial\alpha/\partial\xi$, $\kappa_l=(\pi N\omega_l/n_l^2)\partial\alpha/\partial\xi$, and $\kappa_s=(\pi N\omega_s/n_s^2)\partial\alpha/\partial\xi$. N is the molecular density, ω the angular frequency, m the effective mass for the ξ vibration, and ω_R the resonance frequency. For SBS, $\kappa_{l,s}=\omega_{l,s}/4n_{l,s}^2$ and $\kappa_\xi=\gamma_e^2k_\xi/16\pi\rho_0v_\xi$. γ_e is the coefficient of electrostriction, $k_\xi\simeq k_l+k_s$, ρ_0 the equilibrium mass density, v_ξ the speed of sound, and $\xi_{\rm SBS}=-i\epsilon$, ϵ being the amplitude of the deviation in the dielectric constant. n is the index of refraction. Equation (1) is the 3WRI picture of SRS-SBS processes in the absence of damping of the material excitations.

In the IST formalism of the 3WRI system (1), the Q_i's act as "potentials" in the third-order Zakharov-Manakov scattering eigenvalue problem.[14] For our considerations of SRS and SBS in this Letter, the initial ($t=0$) and the final (after the interactions have ceased) wave packets for the Q_i's will be considered to be nonoverlapping. The individual wave packets are then described[16,18] by the simpler, second-order Zakharov-Shabat (ZS) eigenvalue problem,[19]

$$(\mathbf{I}\partial/\partial\mathbf{x}-\mathbf{U}^{(i)})\mathbf{u}^{(i)}=\mp i\lambda^{(i)}\mathbf{u}^{(i)}.$$

$\lambda^{(i)}$ is the eigenvalue, $\mathbf{u}^{(i)}=(u_1^{(1)},u_2^{(i)})^T$ the eigenvector, and $\mathbf{U}^{(i)}=(q^{(i)},r^{(i)})^T$ the ZS potentials, where

$$q^{(m)}=i\gamma_n\gamma_pQ_m^*/[(-1)^{m+1}\beta_{mn}\beta_{mp}]^{1/2},$$
$$r^{(m)}=(-1)^{m+1}\gamma_n\gamma_pq^{(m)*},$$
$$\lambda^{(m)}=(\zeta/2)(-1)^{m+1}\beta_{np},$$
$$\beta_{np}=v_p-v_n,\quad m,n,p \text{ cyclic}.$$

The Stokes growth and the medium polarization are determined by the ZS scattering data for envelopes 1 and 2, respectively. For nonoverlapping wave packets, the final scattering data are related to the initial scattering data obtained by scattering a plane wave off the envelopes. In SRS and SBS, $r^{(2)}=+q^{(2)*}$ and the ZS problem for the middle envelope is Hermitian and no soliton exchange from the middle to the other two envelopes takes place.[16,18] The scattering data then correspond to the continuous spectrum which is characterized by the "reflection coefficient" $\rho(\lambda)=(b/a)\lambda$, a and b being the scattering coefficients in the Jost-function expansions. More precisely, it is customary[18] to express the wave packets in terms of the "density of radiation" Γ defined as

$$\Gamma(\lambda)=[1\mp|\rho(\lambda)|^2]^{\mp}-1,\quad r=\pm q^*.$$

Solving the ZS problem for square wave packets, the following single-pulse expressions are obtained for $\Gamma_f^{(1)}$ and $\Gamma_f^{(2)}$:

$$\Gamma_f^{(1)}=[\Gamma_0^{(1)}(1+\Gamma_0^{(3)})+\Gamma_0^{(2)}\Gamma_0^{(3)}+2(1+\Gamma_0^{(3)})\mathrm{Re}\Sigma_0]/[1+\Gamma_0^{(2)}],\tag{2}$$

$$\Gamma_f^{(2)}=[\Gamma_0^{(1)}\Gamma_0^{(3)}+(1+\Gamma_0^{(3)})\Gamma_0^{(2)}+2(1+\Gamma_0^{(3)})\mathrm{Re}\Sigma_0]/[1+\Gamma_0^{(1)}],\tag{3}$$

$$\Sigma_0=-b_0^{(1)}b_0^{(2)}b_0^{(3)}/a_0^{(1)}a_0^{(3)},\qquad |\Sigma_0|^2=\Gamma_0^{(1)}\Gamma_0^{(2)}\Gamma_0^{(3)}/D_4.\tag{4}$$

Subscripts 0 and f denote the initial and the final envelopes, respectively. For square envelopes, $\Gamma_0^{(3)}$ and $\Gamma_0^{(2)}$ are given by

$$\Gamma_0^{(3)}=A_3^2G(\lambda_3^2l_3^2-A_3^2),\qquad \Gamma_0^{(2)}=A_2^2G(4\lambda_2^2l_2^2-A_2^2).\tag{5}$$

The dimensionless "area" A is the integral of the ZS potential $q(x)$ over the pulse length l, and $G(z)=\sinh^2(|z|)^{1/2}/|z|$ if $z\le 0$ and $\sin^2(z)^{1/2}/z$ if $z\ge 0$. For large A, $G\simeq\frac{1}{4}\exp(2A)$ for $0<\lambda<A/l$, whereas Γ is bounded by A^2 if $\lambda>A/1$. Moreover, $q,r\subset L_1$, $x\subset R$.

The single-pulse Stokes growth is determined from Eq. (2) and the polarization from Eq. (3). In QSSS with a repetitive pump pulse train, only $\Gamma_f^{(2)}$ changes with successive pulsing; $\Gamma_0^{(3)}$ and $\Gamma_0^{(1)}$ remaining unaltered. The polarization after the nth pulse is then obtained by iterating Eq. (3) n times, $\Gamma_0^{(1)}$, $\Gamma_0^{(2)}$ (single pulse), and $\Gamma_0^{(3)}$ being given. The Stokes growth at the end of the nth pulse is then obtained by replacing $\Gamma_0^{(2)}$ in Eq. (2) by its iterated value. We now apply these results to SRS and SBS in the QSS regime.

SRS in the QSS regime—. For SRS the middle envelope is zero at $t=0$. The phase of the quantity $\mathrm{Re}\Sigma_0$ in Eqs. (2) and (3) depends primarily on λ and not on Γ and for SRS, which is usually initiated by self-focusing, $\Gamma_0^{(1)}\sim(10^{-4}\text{–}10^{-5})\Gamma_0^{(3)}$.[20] These considerations along with Eq. (4) permit neglecting the terms involving $\mathrm{Re}\Sigma_0$ in the first approximation. The nth iteration then yields the following expression for $\Gamma_f^{(2)}$:

$${}^{(n)}\Gamma_f^{(2)}=[\Gamma_0^{(1)}/(1+\Gamma_0^{(1)})][(1+\Gamma_0^{(3)})^n-1],\tag{6}$$

in the absence of damping of the material excitation. However, since there can be no scattering in between pulses, then for interaction times shorter than damping time, damping can be introduced phenomenologically, in which case Eq. (6) is modified as

$${}^{(n)}\Gamma_f^{(2)}=\frac{h\{[D_4\exp(-2t_c/T)]^n-1\}}{D_4\exp(-2t_c/T)-1}.\tag{7}$$

h is the quantity within the first square brackets of Eq. (6), t_c the pulse repetition time, and T the amplitude damping rate. Equation (7) immediately implies that if

$T \ll t_c$, the QSS condition is reached after only a single pulse; that is, $n=1$. Physically, this means that at the end of t_c the material excitations are already in equilibrium so that each pulse in the train encounters identical gain. On the other hand, if $T \gg t_c$, Eq. (7) indicates a larger value for n. Indeed Eq. (2) shows that QSS is attained only if $\Gamma_f^{(2)} \gg 1$ in which case $\Gamma_f^{(1)} \approx \Gamma_0^{(3)}$. For a pump around threshold, this corresponds to 100% reflection. Each of these conclusions of the IST analysis is fully borne out by numerical computations.[11]

In order to demonstrate how well IST estimates the number of pulses required for establishing the QSS regime, the Stokes growth, and the medium polarization, let us apply the above results to the numerical experiments of Ref. 11, where $I_l = 20$ MW/cm^2, FWHM equals 9 mode spacings of 313.84 MHz each, yielding a pulse duration of 0.1269 ns or a pulse length l_l of 2.488 cm (assuming $n=1.53$), $t_c=\pi$ ns, $T=10$ ns, and $\Gamma_0^{(1)} = 10^{-4}\Gamma_0^{(3)}$. The laser pulse area A_3 can be expressed in terms of the Raman cross section as

$$A_3^2 = I_l l_l^2 (2\pi^2 c/nh\omega_l^3) N\, d\sigma/d\Omega\,. \tag{8}$$

For a liquid such as CS_2 (for which[21] $N\, d\sigma/d\Omega \sim 7.55\times10^{-8}$ cm^2/sr) pumped by a ruby laser ($\omega_l = 2.7$ THz), Eqs. (8) and (5) yield a value of 3.37 for $\Gamma_0^{(3)}$. Equation (6) yields $n \simeq 8$ for $\Gamma_f^{(2)}$ above 5. If, on the other hand, Eq. (7) is used, then $n \approx 14$. The polarization in this case increases in a sawtooth fashion eventually reaching saturation. These are in excellent agreement with the results obtained by extensive numerical computations of Ref. 11.

SBS in the QSS Regime—. We now consider SBS and compare the IST results with the experimental observations of Ref. 12 in N_2 gas at 100 bars using a Nd:YAlG (1.06 μm, $\omega_l = 1.776$ THz) laser with a pulse width of 0.9 ns (pulse length 26.4 cm). For SBS, $\Gamma_0^{(1)} = 0$ and the laser area A_3 is

$$A_3^2 = I_l l_l^2 \gamma_e^2 \omega_l^2 / 8\rho_0 v_p c^4\,. \tag{9}$$

$\gamma_e = \rho_0(\partial\epsilon/\partial\rho)_T$. For N_2 we use[22] $n=1.0232$, $\rho_0 = 9.44\times10^{-2}$ g/cm^3, $v_p = 339$ m/s, and a value of 30 GW/cm^2 for I_l which is slightly below the breakdown value of 35 GW/cm^2. Equation (9) then yields a value of 7.7 for A_3.

The acoustic pulse area A_2 for SBS is given by

$$A_2^2 = \omega_l^2 l_2^2 \langle\epsilon^2\rangle / 16 n^2 c^2\,. \tag{10}$$

Its evaluation is relatively more difficult due to the factor $\langle\epsilon^2\rangle$. However, an order-of-magnitude estimate can be obtained from thermodynamical considerations[22] according to which $\langle\epsilon^2\rangle = \gamma_e^2 v / V_{sc} N_A$, where v is the gram-mole volume, $N_A = 6.023\times10^{23}$ is Avogadro's number, and V_{sc} the scattering volume. From the experimental data of a focusing lens of 1-m focal length and an energy of 28 mJ in a pulse of duration 0.9 ns, the spot size is estimated as 0.03 cm. Taking v to be 0.3 liter yields $\langle\epsilon^2\rangle \sim 10^{-22}$ for cells of a few tens of centimeters. From Eq. (10), the pulse area A_2 is obtained as $\sim 10^{-5}$–10^{-6}.

In the experiments of Ref. 12, a 28-mJ pulse (intensity below the breakdown value of 35 GW/cm^2) was followed by a 14-mJ pulse, with a final 6.2-mJ pulse, resulting in QSS with almost full conversion. From Eq. (9) the corresponding A_3 values are 7.7, 5.5, and 3.6, respectively. From iterations of Eq. (3), ${}^{(1)}\Gamma_f^{(2)} \sim 0.8\times10^{-5}$, ${}^{(2)}\Gamma_f^{(2)} \sim 0.12$, and ${}^{(3)}\Gamma_f^{(2)} \sim 39$ demonstrating that QSS is reached with the third pulse with almost 100% conversion. The IST estimates are again in agreement with the experimental observations.

It is interesting to calculate the laser pulse area A_{3t} for threshold, namely, $A_{3t} = \sinh^{-1}(1/A_2)$.[18] For $A_2 \sim 10^{-5}$–10^{-6}, A_{3t} is roughly 13. This is approximately the combined sum of the areas of the first two iterations and shows that the reflection coefficient underwent a jump at the end of the second pulse. This is exactly the experimental observation of Ref. 12. Similar conclusions also hold for the SRS process discussed earlier.

In conclusion, stimulated Raman and Brillouin scattering in the quasi-steady-state regime are analyzed by the inverse-scattering transform considering nonoverlapping square wave packets and the second-order Zakharov-Shabat eigenvalue problem as the scattering problem. The application of the IST formalism readily determines the number of pulses required for the establishment of the QSS condition, the Stokes growth, and the development of the medium polarization. These results cannot be obtained except by extensive numerical computations which, unfortunately, yield no information as to the fundamental determination of these quantities. The agreement of the IST results with experimental observations and numerical computations is excellent.

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