Influence of group-velocity mismatch and inertia of optical nonlinearity on slow-light effects in stimulated inelastic scattering of light

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(Received 3 May 2007; revised manuscript received 9 July 2007; published 20 September 2007)

Group-velocity mismatch between the pump and Stokes pulses and the inertia of optical nonlinearity are shown to modify scenarios of slow-light evolution of laser pulses accompanying stimulated Raman and Brillouin scattering (SRS and SBS) of light. In the transient regime of SRS and SBS, the inertia of nonlinear polarization of a medium shifts the peak of an amplified Stokes field envelope relative to the input Stokes pulse. This shift is sensitive to the shape of input laser pulses and is controlled by the SRS and/or SBS gain, the damping time of the optical phonon wave, and the delay time between the input pump and Stokes pulses. In the regime where both the inertia of nonlinear polarization of an SRS medium and group-delay effects are significant, stationary modes of the Stokes field may exist in the frame of reference propagating with the group velocity of the pump field. The maximum group delay attainable for such modes of the Stokes field is limited, similar to the steady-state regime, by the ratio of the steady-state SRS gain to the Raman gain bandwidth.

DOI: 10.1103/PhysRevA.76.033825 PACS number(s): 42.65.Wi

I. INTRODUCTION

Slowing light down to group velocities substantially lower than the speed of light in vacuum c is one of the most significant recent achievements of optical science [1–3]. While the fundamental research has been gaining a new momentum due to slow-light generation using electromagnetically induced transparency [4], coherent population oscillations [5], photonic-crystal waveguides [6–8], demonstration of optically controlled laser pulse delay and advancement through stimulated Raman [9,10] and Brillouin [11–14] scattering (SRS and SBS) opens new horizons in telecommunication and optical information technologies [15–17], allowing the development of critical components for bit-level synchronizers, signal processors, and tunable data buffers in an all-fiber or silicon-on-insulator-chip format.

Most of the recent experiments on slow light in optical fibers and silicon chips have employed the steady-state regimes of SRS and SBS, where the optically tunable delay and advancement of laser pulses can be adequately described in terms of a group index dramatically increased within the Raman or Brillouin gain band [10,11,16]. In view of the on-going search for compact systems and efficient methods capable of providing higher bit-rate data streams, the extension of SRS and/or SBS-based slow-light techniques to shorter light pulses would be of interest. As shown by experiments reported by Okawachi *et al.* [10], silicon chips pumped by 7-ps laser pulses can serve as efficient and compact SRS-based tunable pulse delays, offering significant advantages over SRS-based fiber-optic delay lines. This tendency of using shorter laser pulses and materials with narrow gain bandwidths in search for higher SRS and/or SBS gains and higher bit rates of data transmission calls for the development of an adequate formalism for the description of slow- and fast-light effects beyond the applicability range of the steady-state treatment, where the laser pulse width τ is assumed to be much greater than the damping time of an optical phonon wave involved in the SRS and/or SBS process. In the case of silicon, for example, the phonon bandwidth $\Gamma/2\pi$ is typically estimated as approximately 105 GHz, suggesting that deviations from steady-state scenarios of stimulated light scattering can show up for laser pulses with pulse widths in the order of a few picoseconds.

A powerful approach to the analysis of Raman-type effects in pulse-propagation dynamics in dispersive nonlinear media is based on the numerical solution of the generalized nonlinear Schrödinger equation (GNSE). This framework has been originally developed to adequately model group-velocity and high-order dispersion effects, Kerr and Raman nonlinearities, absorption, as well as shock-wave formation in the nonlinear evolution of short-pulse light fields in optical fibers. In recent work, the GNSE-based approach has been extended to include two-photon absorption and absorption due to free carriers in semiconductors and augmented by rate equations governing free-carrier generation, resulting in the development of a powerful framework for a theoretical analysis of Raman-mediated amplification of laser fields in silicon-wire waveguides (see Refs. [18–20] for a comprehensive and illuminating overview). While the rate equations for free-carrier number density included in this modified GNSE-based framework help to provide a self-consistent description of transient effects related to free-carrier absorption, the Raman nonlinearity is still included in this formalism phenomenologically, through the use of a damped oscillator model of the Raman response function, corresponding to a Lorentzian spectral profile of the Raman nonlineaer susceptibility in the frequency domain.

Here, we focus on transient effects in stimulated inelastic scattering of light not included in the GNSE-based framework, showing that the inertia of optical nonlinearity may have a significant influence on slow-light phenomena in stimulated inelastic scattering of light, such as SRS and SBS. The plan of this paper is as follows. In Sec. II, we examine SRS and/or SBS-induced slow-light effects for group-index-mismatched Stokes and Raman fields. In Sec. III, we consider slow- and fast-light phenomena in SRS and/or SBS media with inertial nonlinear polarization in the regime where the laser pulse width is less than the damping time of the phonon wave. Section IV discusses slow-light effects in stationary modes of the Stokes field, existing in the regimes of SRS where both the inertia of nonlinear polarization and group-delay effects are significant. The main results of this analysis are briefly summarized in the Conclusion.

II. SLOW-LIGHT EFFECTS IN GROUP-VELOCITY-MISMATCHED STIMULATED SCATTERING

Our analysis is based on a standard set of coupled equations for the slowly varying envelopes of the pump and Stokes fields, $A_p \equiv A_p(t,z)$ and $A_s \equiv A_s(t,z)$, and the vibrational coordinate $Q \equiv Q(t,z)$, representing the envelope of an optical phonon wave [21,22]:

$$\frac{1}{u_p}\frac{\partial A_p}{\partial t} + \frac{\partial A_p}{\partial z} = -i\gamma_p Q A_s, \tag{1}$$

$$\frac{1}{u_s}\frac{\partial A_s}{\partial t} + \frac{\partial A_s}{\partial z} = -i\gamma_s Q^* A_p, \tag{2}$$

$$\frac{\partial Q}{\partial t} + (\Gamma/2 + i\Delta\omega)Q = -i\gamma_Q A_s A_p^*, \tag{3}$$

where u_p and u_s are the group velocities of the pump and Stokes pulses, respectively, Γ is the damping rate of the phonon wave, which defines the SRS and/or SBS gain bandwidth, $\gamma_p = 2\pi N\omega_p c^{-1} n_p^{-1}\partial\alpha/\partial Q$, $\gamma_s = 2\pi N\omega_s c^{-1} n_s^{-1}\partial\alpha/\partial Q$, $\gamma_Q = 2^{-1}N\omega_Q^{-1}\partial\alpha/\partial Q$, α is the polarizability, N is the number density of Raman-active species, ω_s and ω_Q are the central frequencies of the Stokes field and the phonon wave, respectively, $\Delta\omega = \omega_p - \omega_s - \Omega$, Ω is the central frequency of the Raman gain band, and n_p and n_s are the refractive indices at the frequencies ω_p and ω_s. In what follows, Eqs. (1)–(3) and their solutions will be mainly discussed in the context of forward SRS. However, these equations are directly applicable for the analysis of the SBS process and can be readily modified to describe backward inelastic scattering of light [23–26].

For media with broad Raman lines, where the pulse width $\tau \gg \Gamma^{-1}$, deviations in light pulse evolution from the steady-state SRS amplification scenario are dominated by group-delay effects. To examine this regime of SRS-induced slow-light generation, we set $\partial Q/\partial t = 0$ in Eq. (3) and reduce Eqs. (1) and (2) to

$$\frac{1}{u_p}\frac{\partial A_p}{\partial t} + \frac{\partial A_p}{\partial z} = -\frac{g_{0p}}{2(1+2i\Delta\omega/\Gamma)}|A_s|^2 A_p, \tag{4}$$

$$\frac{1}{u_s}\frac{\partial A_s}{\partial t} + \frac{\partial A_s}{\partial z} = \frac{g_{0s}}{2(1-2i\Delta\omega/\Gamma)}|A_p|^2 A_s, \tag{5}$$

where $g_{0p} = 4\gamma_Q\gamma_p/\Gamma$ and $g_{0s} = 4\gamma_Q\gamma_s/\Gamma$.

In the approximation of undepleted pump, $A_p(\eta_p,z) = A_p(\eta_p,0) \equiv A_p(\eta_p)$, where $\eta_p = t - z/u_p$, the solution for the Stokes field is given by

$$A_s(t,z) = A_s(\eta_s,0)\exp\left[\frac{g_{0s}}{2(1-2i\Delta\omega/\Gamma)}\int_0^z |A_p(\eta_s+\xi z')|^2 dz'\right], \tag{6}$$

where $\eta_s = t - z/u_s$ and $\xi = u_s^{-1} - u_p^{-1}$.

The SRS intensity gain is thus given by

$$G_s(t,z) = \frac{g_{0s}}{(1+4\Delta\omega^2/\Gamma^2)}\int_0^z |A_p(\eta_s+\xi z')|^2 dz', \tag{7}$$

and the SRS-induced phase shift of the Stokes pulse is written as

$$\Phi_s(t,z) = \frac{\Delta\omega g_{0s}}{\Gamma(1+4\Delta\omega^2/\Gamma^2)}\int_0^z |A_p(\eta_s+\xi z')|^2 dz'. \tag{8}$$

The related group delay of the Stokes field is

$$\tau_g(t,z) = \frac{\partial\Phi(t,z)}{\partial\omega_s} = \frac{g_{0s}}{\Gamma}S(\Delta\omega)\int_0^z |A_p(\eta_s+\xi z')|^2 dz', \tag{9}$$

where

$$S(\Delta\omega) = \frac{(1-4\Delta\omega^2/\Gamma^2)}{(1+4\Delta\omega^2/\Gamma^2)^2}.$$

When the group velocities of the pump and Stokes fields are matched so that $\xi \ll \tau_p/l$, where τ_p is the pump pulse width and l is the length of the Raman-active medium, Eq. (9) is reduced to

$$\tau_g = S(\Delta\omega) g_{0s}|A_p|^2 z, \tag{10}$$

recovering the well-known result [16] for the SRS-induced change in the group index

$$\Delta n_g = S(\Delta\omega) c g_{0s}|A_p|^2. \tag{11}$$

The peak SRS intensity gain in this regime at $\Delta\omega = 0$ is given by the standard expression for the steady-state SRS gain:

$$G_0 = \frac{4\gamma_Q\gamma_s|A_p|^2 z}{\Gamma}. \tag{12}$$

With $\xi \neq 0$, group-delay effects decrease (Fig. 1) the SRS-induced group delay relative to τ_g values attainable when the group velocities of the pump and Stokes fields are matched (e.g., by using a waveguide structure with an appropriately designed dispersion profile).

Notably, while in the stationary SRS regime with $\xi = 0$, the group delay of the Stokes field is given by Eq. (10), scaling as $\propto |A_p|^2 l$ with the pump intensity $|A_p|^2$ and the interaction length l, in the regime where group-delay effects become significant ($\xi \neq 0$), the linear scaling of the group delay τ_g with the interaction length fails. Thus, in contrast to slow-light effects in stationary ($\xi = 0$) SRS and SBS, which can be adequately described in terms of a group index change defined by Eq. (11), in the case of $\xi \neq 0$, an SRS and/or SBS-induced change in the group index is no longer meaningful as a parameter related to the entire medium, but can be introduced only as a local, z-dependent factor.

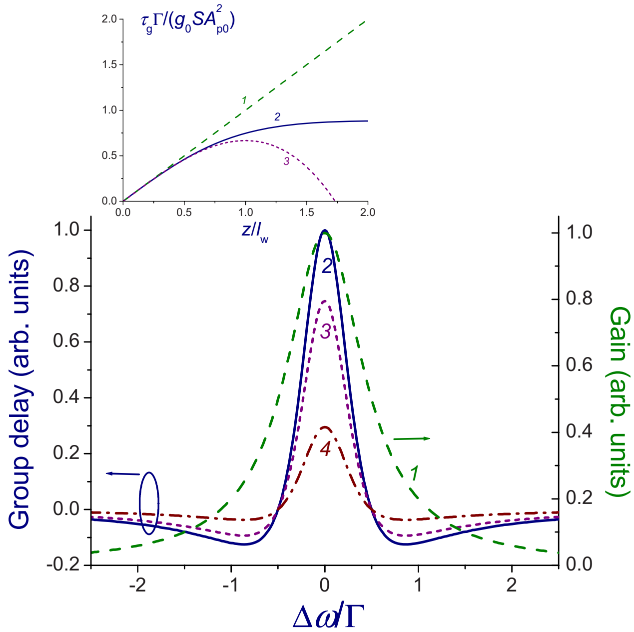

FIG. 1. (Color online) The peak SBS and/or SRS gain $G_s(0,l)/(g_0A_{p0}^2l)$ for $\xi=0$ (1) and the group delay $\tau_g(0,l)\Gamma/(g_0A_{p0}^2l)$ for $\xi=0$ (2), τ_p/l (3), and $3\tau_p/l$ (4) as functions of the frequency detuning $\Delta\omega/\Gamma$ from the central frequency Ω of the SRS and/or SBS gain band. The inset shows the normalized group delay $\tau_g(0,z)\Gamma/(g_0SA_{p0}^2)$ as a function of z/l (l is the interaction length) for a Gaussian pulse with $\xi=0$ (1) and $\xi=\tau_p/l$ (2, 3) calculated by using Eq. (9) (lines 1, 2) and Eq. (15) (line 3).

For a broad class of pulse shapes, the pump field can be represented as

$$|A_p(\eta_p)|^2 = A_{p0}^2 f(-\eta_p^2/\tau_p^2), \tag{13}$$

where A_{p0} is the peak value of the pump field and $f(\eta_p)$ is its temporal profile, which is assumed to reach its maximum at $\eta_p=0$, $f(0)=1$. A formal Taylor-series expansion of the integrand in Eqs. (6)–(9) at the peak of the pump field ($\eta_p=0$), corresponding to the maximum SRS gain, then yields

$$|A_p(\eta_s+\xi z')|^2 \approx A_{p0}^2\left[1-\frac{\xi^2}{\tau_p^2}f'(0)(z'-z)^2\right], \tag{14}$$

where $f'(0)=df(\zeta)/d\zeta|_{\zeta=0}$.

Substituting Eq. (14) into Eq. (9) and performing the integration in the resulting expression, we arrive at

$$\tau_g(0,z) = \frac{g_{0s}}{\Gamma}S(\Delta\omega)A_{p0}^2 z\left[1-\frac{\xi^2}{3\tau_p^2}f'(0)z^2\right]. \tag{15}$$

Group-delay-induced pulse walk-off effects can thus substantially decrease the SRS-induced group delay of the Stokes pulse within a characteristic spatial scale

$$l_g = \sqrt{\frac{3}{f'(0)}}\frac{\tau_p}{|\xi|}. \tag{16}$$

For a Gaussian pump pulse, $f(\zeta)=\exp(\zeta)$, $f'(0)=1$, Eq. (16) is reduced to

$$(l_g)_G = \sqrt{3}\frac{\tau_p}{|\xi|}. \tag{17}$$

For copropagating pump and Stokes fields, $|\xi|\approx|\beta_2\Omega|$, where $\beta_2=\partial^2\beta/\partial\omega^2$, β being the propagation constant of the relevant waveguide mode, and the characteristic spatial scale $(l_g)_G$ can be estimated as

$$(l_g)_G \approx \sqrt{3}\frac{\tau_p}{|\beta_2|\Omega}. \tag{18}$$

For counterpropagating pump and Stokes fields, the above-defined group-velocity mismatch parameter ξ should be replaced by $\zeta=u_s^{-1}+u_p^{-1}\approx 2n_g/c$, where n_g is the group index of the pump field. In this regime, group-delay-induced pulse walk-off effects start to noticeably modify a slow-light evolution of laser pulses within a spatial scale

$$(l_g)_G \approx \frac{\sqrt{3}}{2}\frac{c\tau_p}{n_g}. \tag{19}$$

For forward SRS in a silicon waveguide with $|\beta_2| \approx 1\ \mathrm{ps}^2/\mathrm{m}$ and $\Omega/2\pi\approx 15.6$ THz, Eq. (18) yields $(l_g)_{\mathrm{G,SRS}} \approx 1.8$ cm for $\tau_p\approx 1$ ps. For backward SBS in a silica fiber, on the other hand, we use Eq. (19) with $n_g\approx 1.46$ to find $(l_g)_{\mathrm{G,SBS}}\approx 17$ cm for $\tau_p\approx 1$ ns.

III. TRANSIENT STIMULATED SCATTERING OF LIGHT IN MEDIA WITH INERTIAL NONLINEAR POLARIZATION

To demonstrate significant general tendencies of pulse delay and advancement in the transient regime of stimulated light scattering, we employ the analytic solution to Eqs. (2) and (3) in the approximation of an undepleted pump, $A_p(\eta_p,z)=A_p(\eta_p,0)\equiv A_p(\eta_p)$, at the center of the SRS and/or SBS gain band, $\Delta\omega=0$, with an initial condition $Q(z-u_Qt)=0$ for $t\to-\infty$ (u_Q being the group velocity of the phonon wave), corresponding to no phonon wave in the medium before the turn-on of the laser field [23,24]:

$$Q^*(\eta_s,\sigma) = i\gamma_Q\int_{-\infty}^{\eta_s}\exp\left[-\frac{\Gamma}{2}(\eta_s-\eta')\right]A_p^*(\eta')A_s(\eta',0)I_0 \times(2\{\gamma_Q\gamma_s\sigma[W(\eta_s)-W(\eta')]\}^{1/2})d\eta', \tag{20}$$

$$A_s(\eta_s,z) = A_s(\eta_s,0)+(\gamma_Q\gamma_s z)^{1/2}A_p(\eta_s) \times\int_{-\infty}^{\eta_s}\exp\left(-\frac{\Gamma}{2}(\eta_s-\eta')\right)\frac{A_p^*(\eta')A_s(\eta',0)}{[W(\eta_s)-W(\eta')]^{1/2}} \times I_1(2\{\gamma_Q\gamma_s\sigma[W(\eta_s)-W(\eta')]\}^{1/2})d\eta', \tag{21}$$

where $\sigma=z-u_Qt$, $W(x)=\int_{-\infty}^{x}|A_p(\zeta)|^2d\zeta$, and $I_i(x)$ is the ith-order modified Bessel function.

In Figs. 2(a) and 2(b), we illustrate the Stokes pulse amplification dynamics for an input field defined as a pair of Gaussian light pulses $E_{1,2}(\theta)=a_{1,2}\exp(-\theta^2/\tau^2)$ with the same pulse width τ and $a_2/a_1=0.01$, serving as a pump and Stokes fields in SRS. Temporal envelope of the pump pulse is shown by curves 1 in Figs. 2(a) and 2(b). For $\Gamma\tau=10$, the

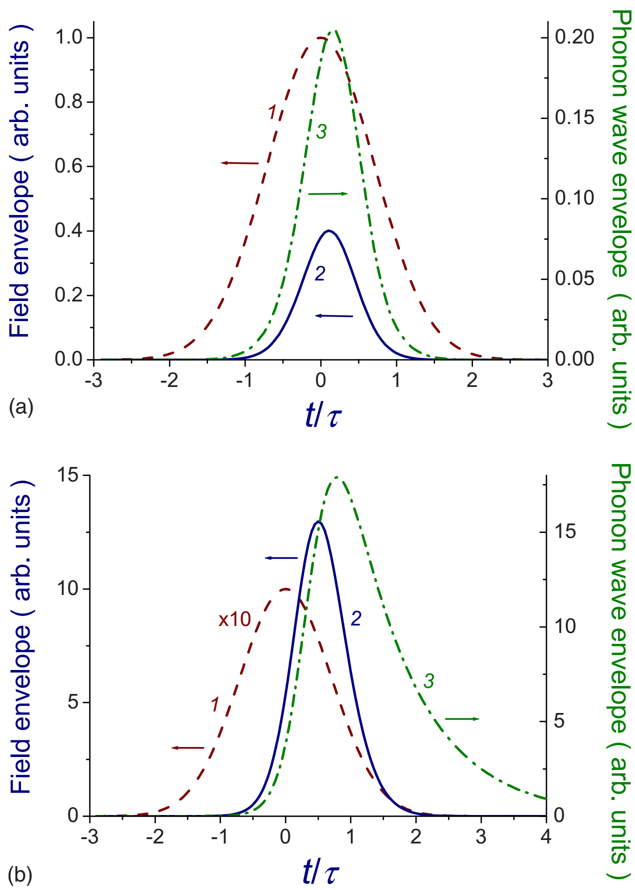

FIG. 2. (Color online) Temporal envelopes of the pump (1) and output Stokes (2) pulses and the phonon wave Q (3) for $\Gamma\tau=20$ (a) and 2 (b) with $G_0=50/\Gamma\tau$ and $a_2/a_1=0.01$. Note a magnifying factor of 10 for the envelope of the pump pulse in Fig. 2(b).

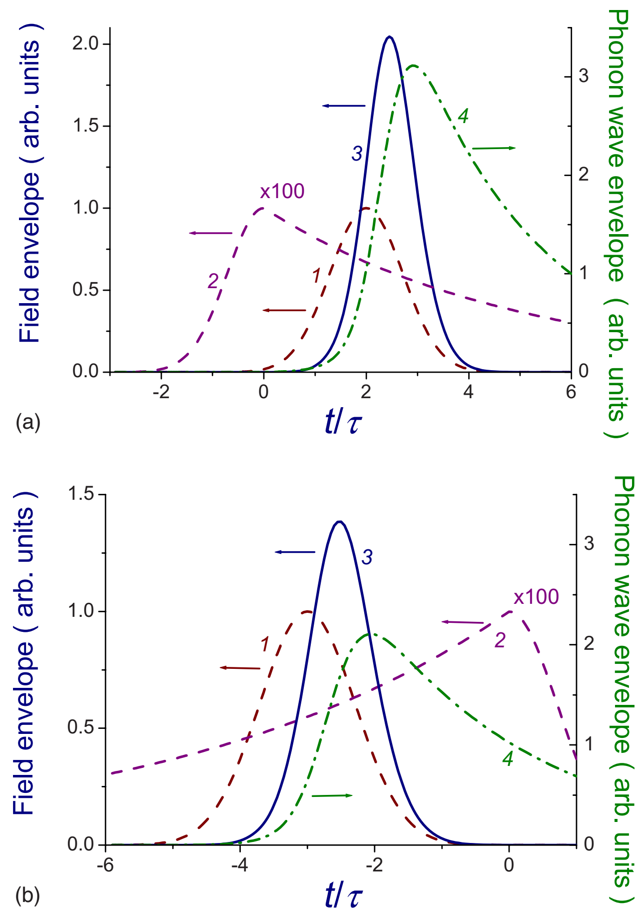

FIG. 3. (Color online) Temporal envelopes of the pump (1), input Stokes (2), and output Stokes (3) pulses and the phonon wave Q (4) for $\Gamma\tau=0.8$, $G_0=25$, $a_2/a_1=0.01$. Delay time of the pump pulse is 2τ (a) and -3τ (b). Note a magnifying factor of 100 for the envelope of the input Stokes pulse.

phonon wave envelope Q [curve 2 in Fig. 2(a)] closely follows the envelopes of the input pulses, leading to virtually no delay between the input pulses and the amplified Stokes field [curve 3 in Fig. 2(a)]. By contrast, in the regime where $\Gamma\tau=1$, the buildup of the phonon wave Q [curve 2 in Fig. 2(b)] lags behind the input field, modifying the field envelope of the amplified Stokes pulse [curve 3 in Fig. 2(b)] and delaying the peak of the amplified Stokes field with respect to the input Stokes pulse.

In Figs. 3(a) and 3(b), we demonstrate that laser pulse delay and advancement in SRS and SBS media can be enhanced by pulse shaping. Here, we define the input pump field as an ultrashort Gaussian pulse with a pulse width τ, choosing the input Stokes field in the form of a pulse with an extended trailing [Fig. 3(a)] or leading [Fig. 3(b)] edge and a maximum shifted in time with respect to the peak of the pump pulse. The rise time of the Stokes pulse with an extended trailing edge [Fig. 3(a)] and the falloff time of the Stokes pulse with an extended leading edge [Fig. 3(b)] are set equal to the pulse width of the pump pulse τ. An optical phonon wave can be stimulated only in the presence of both the stokes field and an ultrashort pump pulse [curves 4 in Figs. 3(a) and 3(b)], allowing amplification of the Stokes field [cf. curves 2 in Figs. 3(a) and 3(b)] within a limited time gate, defined by the ultrashort pump pulse. As a result, the amplified Stokes pulse can be delayed by a few pulse rise and/or falloff times τ with respect to the input Stokes field.

IV. GROUP-VELOCITY-MISMATCHED TRANSIENT STIMULATED RAMAN SCATTERING

In the regime of SRS where both the inertia of nonlinear polarization of a medium and group-delay effects are significant, i.e., $\Gamma\tau<1$ and $\xi l/\tau_p>1$, normal dispersion ($u_p<u_s$) tends to delay the pump relative to the Stokes pulse, pushing the pump field to the region where molecular vibrations have been just excited through the joint action of the pump and Stokes fields. In this situation, as highlighted by Carman *et al.* [23], more Stokes radiation can be generated through a more efficient interaction of the pump with the phonon wave. As shown by Akhmanov *et al.* [25], Eqs. (2) and (3) with an initial condition for an undepleted pump field $A_p(\eta)=A_{p0}[\cosh(\eta/\tau_p)]^{-1}$ allow a stationary solution for the Stokes field for $z>l_w\equiv\tau_p/|u_p^{-1}-u_s^{-1}|$ in the retarded frame of reference (η, z), moving with the group velocity of the pump pulse,

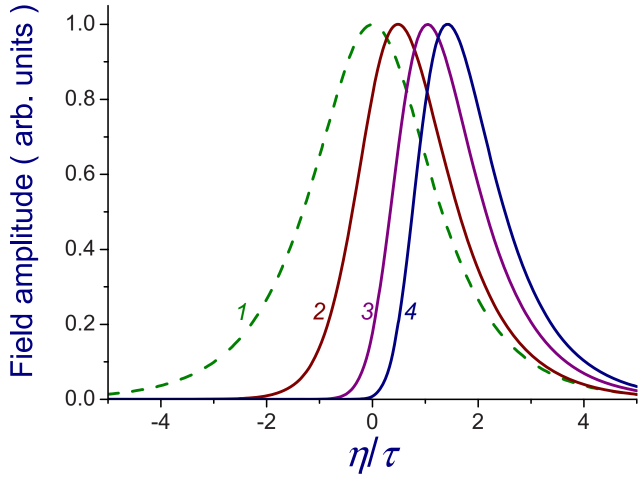

FIG. 4. (Color online) The normalized pump field envelope (1) and stationary Stokes pulses defined by Eq. (22) with $\Gamma\tau_{\mathrm{p}}=0.2$ and $g_{\mathrm{s}}=2$ (3), 5, (4), and 10 (4).

$$A_s(\eta) = A_{s0} \exp\left[\left(g_s - 1 - \frac{\Gamma\tau_p}{2}\right)\frac{\eta}{\tau_p}\right]\left[\cosh\left(\frac{\eta}{\tau_p}\right)\right]^{-g_s}, \tag{22}$$

where

$$g_s = |A_p|\tau_p[\gamma_Q\gamma_s(u_p^{-1} - u_s^{-1})]^{1/2}. \tag{23}$$

The gain of the stationary Stokes mode defined by Eq. (22) is

$$G_s = \frac{2g_s - 1 - \Gamma\tau_2/2}{l_w}. \tag{24}$$

In the strong-pump regime, Eq. (24) yields

$$G_s = 2|A_p|[\gamma_Q\gamma_s(u_p^{-1} - u_s^{-1})]^{1/2}. \tag{25}$$

We now use the solution given by Eq. (22) to examine slow-light effects in group-velocity-mismatched transient SRS. As the Stokes field is stationary in the frame of reference moving with the pump pulse, we can represent the group delay of the Stokes field induced by SRS gain in the considered regime as

$$\tau_g = (u_p^{-1} - u_s^{-1})z. \tag{26}$$

This corresponds to an SRS-induced change in the group index

$$\Delta n_g = c(u_p^{-1} - u_s^{-1}). \tag{27}$$

Physically, the existence of this slow-light stationary Stokes mode becomes possible when the Raman gain leads to a strong localization of the Stokes field on the trailing edge of the pump pulse (Fig. 4). Indeed, as can be seen from Eq. (22), the stationary Stokes mode is well localized in time only as long as

$$g_s > \frac{1}{2} + \frac{\Gamma\tau_p}{4}. \tag{28}$$

Since large group delays reduce, according to Eqs. (22)–(25), the SRS gain, the requirement of Eq. (28) limits the group delay times τ_{g} attainable for the stationary Stokes mode (22). To demonstrate this, we take into consideration that $\tau_{\mathrm{p}}\Gamma < 1$ and neglect the second term on the right-hand side of Eq. (28). Substituting Eqs. (12) and (26) into Eq. (28), we then arrive at

$$\tau_g < \frac{G_0}{\Gamma}. \tag{29}$$

Notably, the same ratio of the steady-state SRS gain G_0 to the Raman linewidth Γ defines the maximum group delay time attainable in the steady-state SRS regime [16,27]. The upper bound of G_0 in steady SRS is determined by the amplification of noise arising through spontaneous Raman scattering. In the transient regime, this limitation on G_0 is loosened, as the transient SRS gain is roughly a factor of $(\tau_{\mathrm{p}}\Gamma)^{-1}$ lower than G_0.

V. CONCLUSION

We have shown in this work that group-velocity mismatch between the pump and Stokes fields in stimulated inelastic scattering of light tends to decrease the SRS and/or SBS-induced group delay of the Stokes pulse. In the transient regime of SRS and SBS, the inertia of nonlinear polarization of a medium shifts the peak of an amplified Stokes field envelope relative to the input Stokes pulse. This shift is sensitive to the shape of input laser pulses and is controlled by the SRS and/or SBS gain, the damping time of the optical phonon wave, and the delay time between the input pump and Stokes pulses. In the regime where both the inertia of nonlinear polarization of an SRS medium and group-delay effects are significant, stationary modes of the Stokes field may exist in the frame of reference propagating with the group velocity of the pump field. The maximum group delay attainable for such modes of the Stokes field is limited, similar to the steady-state regime, by the ratio of the steady-state SRS gain to the Raman gain bandwidth. However, since the transient SRS gain is lower than its steady-state counterpart, the limitation on the group delay of the Stokes field, set by the amplification of noise arising through spontaneous Raman scattering, is significantly loosened relative to the steady-state regime of SRS.

ACKNOWLEDGMENTS

This study was supported in part by the Russian Foundation for Basic Research (Projects No. 06-02-16880, No. 07-02-12175, and No. 07-02-91215), Federal Research Program of the Ministry of Science and Education of Russian Federation (Contract No. 02.513.11.3040), and INTAS (Projects No. 03-51-5037 and No. 03-51-5288). The research described in this publication was made possible in part by financial support (Contract No. RUP2-2695) from the U.S. Civilian Research & Development Foundation for the Independent States of the Former Soviet Union (CRDF).

APPENDIX: APPROXIMATE ANALYTICAL SOLUTIONS TO THE COUPLED EQUATIONS FOR THE SLOWLY VARYING ENVELOPES OF THE STOKES FIELD AND THE PHONON WAVE

In this Appendix, we follow the analysis of earlier work [21–24], providing the derivation and a brief explanation of approximate analytical solutions (6), (20), and (21) to the coupled equations (2) and (3) for the slowly varying amplitudes of the Stokes field $A_s \equiv A_s(z,t)$ and the phonon wave $Q \equiv Q(z,t)$.

In media with broad Raman lines, corresponding to a fast Raman response, $\tau \gg \Gamma^{-1}$, the standard approach to the analysis of Eqs. (1)–(3) involves the natural steady-state approximation $\partial Q/\partial t = 0$, which yields the set of Eqs. (4) and (5) for the envelopes of the pump and Stokes fields A_p and A_s. With an assumption of undepleted pump, $A_p(\eta_p,z) = A_p(\eta_p,0) \equiv A_p(\eta_p)$, the solution to Eq. (5) is given by Eq. (6), which can be verified through a direct calculation of partial derivatives of A_s, as defined by Eq. (6), in t and z:

$$\frac{\partial A_s(t,z)}{\partial z} = -\frac{1}{u_s}\frac{\partial A_s(\eta_s,0)}{\partial \eta_s}\Psi(t,z) + A_s(\eta_s,0)\kappa\Psi(t,z) \times\left(-\frac{1}{u_s}\int_0^z \frac{\partial |A_p(\eta_s+\xi z')|^2}{\partial z}dz' + |A_p(\eta_p)|^2\right) \tag{A1}$$

$$\frac{\partial A_s(t,z)}{\partial t} = \frac{\partial A_s(\eta_s,0)}{\partial \eta_s}\Psi(t,z) + A_s(\eta_s,0)\kappa\Psi(t,z) \times\left(\int_0^z \frac{\partial |A_p(\eta_s+\xi z')|^2}{\partial z}dz'\right), \tag{A2}$$

where

$$\Psi(t,z) = \exp\left(\frac{g_{0_s}}{2(1-2i\Delta\omega/\Gamma)}\int_0^z |A_p(\eta_s+\xi z')|^2 dz'\right),$$

$$\kappa = \frac{g_{0_s}}{2(1-2i\Delta\omega/\Gamma)}.$$

Multiplying Eq. (A2) by u_s^{-1} and adding the resulting expression to Eq. (A1), we recover Eq. (5).

To derive Eqs. (20) and (21), we assume that pump depletion is negligible, $A_p(\eta_p,z) = A_p(\eta_p,0) \equiv A_p(\eta_p)$, and the central frequencies of the pump and Stokes fields are chosen in such a way as to meet the condition of a Raman resonance, $\Delta\omega = 0$. We can then follow the arguments by Carman *et al.* [23] by introducing a new variable

$$W = \int_{-\infty}^{x} |A_p(\zeta)|^2 d\zeta \tag{A3}$$

and making use of the substitutions

$$U_1 = Q^* \exp(\Gamma\eta_s), \tag{A4}$$

$$U_2 = A_p^* A_s \exp(\Gamma\eta_s). \tag{A5}$$

to reduce Eqs. (2) and (3) to the following second-order hyperbolic partial differential equation:

$$\frac{\partial^2 U_i}{\partial \sigma\, \partial W} - \gamma_s\gamma_Q U_i = 0, \tag{A6}$$

where $i=1$, 2.

Solving Eqs. (A4) and (A6) for the phonon wave envelope Q^* subject to the initial condition of no Raman excitation in the medium prior to the laser field, $Q(z-u_Q t)=0$ for $t \to -\infty$, yields Eq. (20). We can then use this expression for Q^* to solve Eqs. (A4)–(A6) for $A_s(\eta_s,z)$ with the Stokes field at the input of the Raman medium defined as $A_s(\eta_s,0)$, recovering Eq. (21).

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