Self-focusing versus stimulated Brillouin scattering of laser pulses in fused silica

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Abstract. The coupling between Kerr-induced filamentation and transient stimulated Brillouin scattering (SBS) is theoretically investigated for nanosecond laser pulses propagating in bulk silica. Power thresholds for beam collapse are evaluated by means of three-dimensional numerical simulations. It is shown that the nonlinear self-focusing of powerful pump waves is able to enhance the Stokes component to high fluence levels even if the incident beam exhibits a broad spectral bandwidth. In contrast, pump pulses with a few tens of picoseconds amplitude modulations drastically inhibit SBS.

The stimulated scattering of light is one of the major topics in nonlinear optics. Although several types of stimulated scattering (Raman, Brillouin and thermal Rayleigh) were already discovered in the 1960s, related topics are still highly active [1]–[4]. Stimulated scattering always involves a pump laser beam and a frequency-shifted scattered wave, coupled by either molecular vibrational transitions (Raman) or acoustic waves (Brillouin). The generation of intense, frequency-shifted radiation through stimulated Brillouin scattering (SBS) is currently used in tunable laser sources, coherent optical communication systems, Brillouin amplifiers and sensors. SBS occurs in a large variety of transparent media and plasmas, from single-mode fibers [5] to all-optical silica devices employed, e.g., in large laser facilities devoted to inertial confinement fusion [6, 7]. In materials with no optical absorption, SBS is mainly driven by the electrostriction strain produced by an intense laser pulse with long enough (nanosecond) durations. This strain excites acoustic waves on which a Stokes wave scatters a significant amount of energy, preferably in the direction opposite to the pump one.
For powerful pump beams, the counterpropagating Stokes wave can convey high enough fluence to cause severe damage and SBS appears as a harmful process that limits the pulse energy of high-power laser sources. The standard approach for reducing SBS is the use of broadband lasers, which works quite well in one-dimensional (1D) geometries and discarded Kerr optical response $[8]–[12]$. It turns out that in full three-dimensional (3D) configurations, however, the coupling between SBS and Kerr nonlinearity becomes a crucial issue. Let us indeed recall that Kerr nonlinearities cause modulational instabilities and can even lead to catastrophic wave collapse at high dimensions once the pump peak power exceeds the self-focusing threshold $[13]$. For understanding the initiation of material damage by powerful lasers, it is necessary to model the interplay between SBS and Kerr nonlinearities in 3D geometries, which has not been addressed so far. Investigations $[14, 15]$ limited to low pump intensities $<20\text{GW cm}^{-2}$, such modeling is not limited to SBS. A similar analysis could be applied to interactions between counterpropagating optical solitons $[16]$ or to stimulated Raman scattering (SRS). It also found promising extensions in slow light propagation mediated by SBS in bulk media $[17, 18]$ and in stimulated Rayleigh–Bragg scattering models $[19]$.

Our investigation is mainly motivated by the unexplained data reported in $[6]$, where high-power single-mode pulses in the nanosecond range initiated not only rear damage by Kerr filamentation in fused silica but also front surface damage through SBS. Multimode broadband pumps, expected to suppress Brillouin scattering, were then observed to noticeably decrease the onset distance of filamentation, which was attributed to a higher Kerr nonlinear index. The goal of this work is to explain these observations and propose alternative solutions to inhibit SBS at laser powers above the critical threshold for self-focusing. Because the decay time of Raman phonons is at least two decades below that of Brillouin phonons excited through nanosecond-long pulses $[12]$, SRS is discarded in the present study. We report on generic behavior resulting from the competition between SBS and SF of intense laser pulses in 3D silica samples. Emphasis is put on pump power thresholds, from which the laser and Stokes pulses are amplified by the SF process. In such regimes, an increase of the laser pump bandwidth to the 100 GHz range through phase modulations is shown to keep SBS partly active and contributes to strongly decreasing the self-focusing distance via the development of modulational instabilities. Alternatively, amplitude modulations profiling the pump pulse into a train of picosecond subpulses do suppress Brillouin scattering.

Figure 1 recalls some basic principles. In a 1D Brillouin-active medium, the pump wave is depleted to the benefit of the Stokes wave along the Brillouin gain length $L_B = 1/g_0I_1(0)$, where $g_0$ and $I_1(0)$ denote the phonon–photon coupling coefficient (or SBS gain factor) and the input pump intensity, respectively. The inset details the mechanisms related to SF in bulk media: a dramatic intensity growth occurs through the continuous increase of the refraction index $n(I) = n_0 + n_2I$, where $n_0$ and $n_2$ are the linear and Kerr nonlinear indices. For an unperturbed Gaussian beam with waist $w_0$ and power $P$ (bottom surface plots), the collapse distance can be estimated by the Marburger formula $z_c \simeq L_M = 0.092L_{\text{diff}}/((\sqrt{P}/P_{\text{cr}} - 0.852)^2 - 0.0219)^{1/2}$ $[20]$, where $L_{\text{diff}} = 2k_0w_0^2$ and $P_{\text{cr}} \simeq 3.72\lambda_0^2/8\pi n_0n_2$ are the diffraction length scale and the critical power for self-focusing that both depend on the laser wavelength $\lambda_0$. At large enough powers, instead, pump pulses may decay into multiple filaments triggered by modulational instability (top surface plots). The whole beam is then expected to collapse at earlier distances, $z_c \simeq L_{\text{SF}} = \lambda_0/2\pi n_2I(0)$ $[21, 22]$. 

New Journal of Physics 12 (2010) 103049 (http://www.njp.org/)
Figure 1. The principle of SBS in a 1D stationary medium (fused silica). The inset details the characteristic wave growths induced by SF, i.e. single filamentation for an unperturbed Gaussian beam ($P = 16 P_{cr}$, solid curve and bottom surface plots) and multiple filamentation for pulses perturbed by a 20% amplitude random noise ($P = 40 P_{cr}$, dashed curve and top surface plots). Note the change in intensity scales as the pulse self-focuses. The multifilamented beam collapses at an earlier distance ($z_c \simeq L_{SF} \simeq 1.7$ cm) than that predicted by the Marburger formula ($L_M \simeq 2$ cm).

Involving the above two processes, our model equations describe the coupling between linearly polarized forward (pump) and backward (Stokes) pulses with an acoustic matter wave created by electrostriction [1]. Their slowly varying envelopes, $U_1$ and $U_2$, respectively, have center frequencies close to $\omega_0 = 2\pi c/\lambda_0 = k_0 c/n_0$ ($c$ is the speed of light in vacuum) and group velocities $k' \simeq k_0/\omega_0$ (in silica $n_0 \simeq 1.47$). The phonon wave has the sound velocity $C_s = 5.97 \times 10^5$ cm s$^{-1}$, wave number $q \simeq 2k_1$ ($\vec{q} = \vec{k}_1 - \vec{k}_2$) and frequency $\Omega_B = C_s q$. The model for the optical fields is derived from Maxwell’s equations, where the polarization vector contains linear dispersion to first order, Kerr-induced self- and cross-phase modulation functions $K_i = (n_2 \omega_0/c)(|U_i|^2 + 2|U_i|^2)$ for $i \neq j = 1, 2$ and electrostriction with elasto-optic coefficient $p_{12} \simeq 0.27$ [2, 5, 14]. Assuming moderate fluences $F_i \equiv \int |U_i|^2 dt < 12$ J cm$^{-2}$, we discard photo-induced plasma generation responsible for further damage to the material. The equations are then expressed as

$$(\partial_z + k' \partial_t) U_1 = \frac{i \nabla^2 U_1}{2k_0} - \frac{g_0}{2} Q U_2 + i K_1 U_1, \quad (1)$$

$$(-\partial_z + k' \partial_t) U_2 = \frac{i \nabla^2 U_2}{2k_0} + \frac{g_0}{2} Q^* U_1 + i K_2 U_2, \quad (2)$$

$$\tau_B \partial_t Q + Q = U_1 U_2^* + N, \quad (3)$$

New Journal of Physics 12 (2010) 103049 (http://www.njp.org/)
where $z$ is the propagation variable, $\nabla^2_\perp = \partial^2_x + \partial^2_y$ is the diffraction operator, and the gain factor $g_0 = 3\hbar n_0^2 p_{\perp 2}/C SC^3 \rho_0 \Gamma_B$ involves the bulk density $\rho_0 = 2.21$ g cm$^{-3}$. The field envelopes $U_i$ have been normalized such that $I_i = |U_i|^2$ is their intensity expressed in W cm$^{-2}$. $Q$ denotes the scaled density fluctuation envelope whose spatial dynamics is discarded ($C_\phi/\Gamma_B < 10 \mu$m) and $\tau_B = 2/\Gamma_B$ is the phonon damping rate. $\Gamma_B$ is the Brillouin linewidth [1], related to the full-width at half-maximum of the Brillouin gain spectrum as $\Delta \nu_B = \Gamma_B/2\pi$. It varies with the pump wavelength as $\Gamma_B = q^2 \Gamma'$, where $\Gamma'$ is the material damping parameter. Consequently, the gain factor $g_0$ barely changes with $\lambda_0$, up to small variations induced by the elasto-optic constant $p_{12}$ linked to the electrostriction coefficient $\gamma\rho$ by the relationship $\gamma\rho \approx n_0^2 p_{12}$ [2, 14]. With $p_{12} = 0.27$ [5], $g_0$ takes the value $g_0 \approx 4.5 \text{ cm GW}^{-1}$. In equation (3), $N$ models a thermally driven Gaussian random noise that initiates SBS at ambient temperature following [10]. Equations (1)–(3) are numerically integrated in full 3D geometry for an input pump with spatial and temporal Gaussian profiles $U_1(z = 0) = \sqrt{I_1(0)} \exp[-(x^2 + y^2)/u_0^2 - t^2/t_0^2]$, initial waist $w_0 = 150 \mu$m and 1/e$^2$ duration $t_0 = 2.12$ ns. The silica sample thickness is $L = 5$ cm. We use a split-step spectral scheme with longitudinal grid spacing $\Delta z = \Delta t/k'$, where $\Delta t$ is the time step. Although our analysis is mainly devoted to UV pulses ($\lambda_0 = 355$ nm, $\Gamma_B = 1.87$ ns$^{-1}$ and $n_2 = 3.6 \times 10^{-16}$ cm$^2$ W$^{-1}$), all the coming features are generic and can be refound for, e.g., infrared pulses ($\lambda_0 = 1064$ nm, $\Gamma_B = 0.2$ ns$^{-1}$ and $n_2 = 2.6 \times 10^{-16}$ cm$^2$ W$^{-1}$), as evidenced at the end of this work.

Equations (1)–(3) describe the interplay between SF and SBS. A simple ordering indicates that the typical length scales for diffraction, self-focusing and SBS obey the inequality $L_B < L_{\text{SF}} \ll L_{\text{diff}}$ for pump intensities above 5 GW cm$^{-2}$. Because for our nanosecond pulses the product $\Gamma_B p_\perp$ is of the order of unity, SBS develops in transient regime, for which the 1D intensity gain is classically evaluated by $G_T \sim 2\sqrt{\Gamma B p}/L_B$ for an undepleted plane-wave pump [9, 12]. On the other hand, the Kerr response, although originally small ($\omega_0 n_2 / g_0 c < 2 \times 10^{-2}$), is expected to cause wave collapse inside the medium at high enough powers $P_\perp \equiv \int P_i d\vec{r}_\perp$. Assuming weak SBS and a stationary regime, the pump blowup requires an input peak power $P_1(0) > P_{\text{cr}}/(1 + 2R)$ [13], where $R \equiv P_2(0)/P_1(0)$ is the SBS power reflectivity. In the high SBS gain and nonstationary case, no analytical expressions are available, so that numerical simulations become necessary.

To start with, figure 2 shows the maximum intensities and partial energies $E_i(z) \equiv \int P_i d\vec{r}_\perp$ of the pump and Stokes waves for peak input powers $P_1(0) = 5 P_{\text{cr}}, 16 P_{\text{cr}}$ and $27 P_{\text{cr}}$ in the UV domain ($\lambda_0 = 355$ nm, $P_{\text{cr}} = 0.35$ MW). The conservation law $E_1(z) - E_2(z)$ is const is fulfilled. At low power, the pump intensity slowly increases and a Stokes wave is smoothly generated inside the material, similarly to the low-intensity regimes investigated in [15]. In contrast, at large powers $P_1(0) > 14 P_{\text{cr}}$, the pump starts to collapse inside the sample. Divergence of the Stokes pulse follows, depending on the noise realization and effective pump duration near the focus. As shown by the dotted curves in figures 2(a) and (c), the Kerr response increases the peak intensity of the two components in the sample. However, the self-focus point $z_c$ of the pump clearly lies beyond Marburger distance ($L_M \leq 3.4$ cm for the two highest powers): the pump energy is rapidly transferred to the Stokes component in the ratio $E_1(z_c)/E_1(0) \simeq 1/\lambda$ (figure 2(d)), which keeps the pump’s nonlinear focus at comparable distances, $z_c \simeq 4$–4.5 cm. Exemplary fluence patterns for the reflected and transmitted pulses are shown in figures 2(b) and (d). Among those, the Stokes fluences approach the damage threshold $[F_2^\text{max}(0) \simeq 11.4$ J cm$^{-2}]$. 

New Journal of Physics 12 (2010) 103049 (http://www.njp.org/)
Figure 2. Peak intensities (a, c) and partial energies (b, d) for 355 nm Gaussian pulses with $w_0 = 150 \mu m$, $t_p = 2.12 \text{ ns}$; (a, b) $P_1(0) = 5 P_{cr}$, (c, d) $P_1(0) = 16 P_{cr}$ (dashed curves) and $P_1(0) = 27 P_{cr}$ (solid curves). Note the change in the intensity scales. Blue (green) curves refer to the pump (Stokes) pulse. The solid lines correspond to the complete model; dotted lines discard the full Kerr response. Insets detail Stokes and pump fluences for (b) the $5 P_{cr}$ and (d) the $27 P_{cr}$ pulse in a $400 \times 400 \mu m^2$ section of the $(x, y)$ plane at $z = 0$ and $z = L$, respectively.

Figure 3. Amplitude spectra of the incident Gaussian pump field at 355 nm for (a) no modulation ($\Delta \nu \approx 0.5 \text{ GHz}$) and (b) a phase modulation with $m = 21$, $\nu_m = 2 \text{ GHz}$ ($\Delta \nu \approx 84 \text{ GHz}$). Note the change of scales.

To attenuate the reflected fluences, broadband pumps with bandwidths $\Delta \nu \gg \Delta \nu_B$ can be employed [11, 23]. Using multimode pumps with mode spacing larger than the Brillouin linewidth results in net decrease of the SBS gain whenever the Stokes modes evolve independently of each other. This requirement is fulfilled if the pump coherence length is small compared with the interaction length, i.e. $\Delta \nu L/c \gg 1$. For this purpose, introducing a phase modulation $U_i \rightarrow M(t) \times U_i$ in the form $M(t) = \exp[im \sin(2\pi \nu_m t)]$ creates a multimode spectrum of $1/e$ bandwidth $\Delta \nu \approx 2 m\nu_m$, where $2m \gg 1$ is the number of modes and $\nu_m$ the modulation frequency. As an example, figure 3 compares the amplitude spectra of our unmodulated Gaussian pulse (figure 3(a)) with its phase-modulated counterpart (figure 3(b)), where $m = 21$ and $\nu_m = 2 \text{ GHz}$. With a large enough number of modes, the SBS gain is then expected to fall to $G_T/\sqrt{2m}$ [23].
Figure 4. (a, b) Power profiles for input (black), transmitted pump (blue) and reflected Stokes (green curve) pulses (a) without and (b) with phase modulation at $16P_{cr}$. Peak intensities of phase-modulated pulses with (c) $16P_{cr}$ and (d) $27P_{cr}$ for $m = 21$ and $\nu_m = 2$ GHz. (e) Corresponding partial energies. (f) Peak intensities for amplitude-modulated beams. Plot styles follow those used in figure 2.

The previous property, however, no longer holds at high powers triggering SF, as evidenced in figure 4. Figures 4(a) and (b) illustrate the power profile of the input, transmitted pump and reflected Stokes pulses, for unmodulated and phase-modulated pumps at $16P_{cr}$. With a phase modulation, the trailing part of the pump strongly fluctuates in time, which decreases the SBS reflectivity ($R \approx 1/6$). Nonetheless, due to the Kerr response, the Stokes intensity sharply increases (figures 4(c) and (d)). Inhibiting SBS through phase modulation leads to a much weaker pump depletion, $E_1(L)/E_1(0) \approx 0.88$, over the sample thickness (figure 4(e)). Consequently, the pump gets more strongly focused by the Kerr nonlinearities. Through space–time couplings, the pump then decays into multiple peaks, which not only self-focus at shorter propagation distances, but also produce a turbulent background that favors the growth of Stokes modes. Here, the reflected fluence can still attain significant levels (e.g. $F_2(0) = 6.7 \, \text{J/cm}^2$).

For comparison, we also tested amplitude-modulated pumps, e.g. $M(t) = \cos[m \sin(2\pi \nu_m t)]$. Surprisingly, both the reflected Stokes intensity and fluence ($\sim 6.5 \, \mu\text{J/cm}^2$) always stay close to zero (figure 4(f)). Backscattering is suppressed, while the pump wave undergoes SF at the expected Marburger distances $L_M = 3.4$ and $2.5 \, \text{cm}$ for $P_1(0) = 16P_{cr}$ and $27P_{cr}$, respectively. The reason is twofold. Firstly, the averaged pump intensity is divided by two in the gain argument. Secondly, such modulations break the pump into pulse trains of short periods $1/m\nu_m \sim 24 \, \text{ps}$, which annihilates the creation of acoustic matter waves. Similar results hold for simpler modulations, i.e. $M(t) = \cos(2 m\pi \nu_m t)$ or $\cos^2(2 m\pi \nu_m t)$.

Figure 5 details intensity profiles in the $(x, t)$-plane for unperturbed and modulated pumps with $P_1(0) = 16 \, P_{cr}$. Close to the nonlinear focus, the pump front is depleted near the instant $t \simeq -1 \, \text{ns}$, from which the Stokes wave emerges and grows at decreasing $z$. Without phase modulation, singly peaked structures are amplified (figures 5(a) and (d)). In the opposite case, the optical fields break up into multiple peaks in space and time, as shown by figures 5(b).
Figure 5. Beam profiles in the $(x, t)$ plane with maximum intensity: (a) $I_1(z = 4.35 \text{ cm})$ without modulation, (b) $I_1(z = 0.13 \text{ cm})$ with phase modulation, (c) $I_1(z = 3.3 \text{ cm})$ with amplitude modulation. Panels (d–f) show the intensities $I_2(0)$.

and (e). The pump and Stokes envelopes are multimode, i.e. $U_i = \sum_n A_{1,n} e^{-i(\omega_n - \omega_0)t} (i = 1, 2)$ [11]. For large frequency deviations, the Stokes modes $A_{2,n} \sim \left[ig_0/2k'(\omega_n - \omega_0)\right]Q^*A_{1,n}$ have limited growth and the Kerr effect rapidly takes over SBS. Under these conditions, modulational instability takes place: perturbations characterized by transverse wave numbers $k_{\perp}^2 < 2n_0n_2\omega_0^2(I_1 + I_2 \pm [(I_1 - I_2)^2 + 16I_1I_2]^{1/2})/c^2$ can exponentially increase and lead to pulse breakup [24]. In contrast, with an amplitude modulation, acoustic waves have no time to form. At leading order, equation (1) reduces to $(\partial_z + k'\partial_t)I_1 \sim -g_0\text{Re}(QU_1^*U_2) \rightarrow 0$. The rapid intensity variations preserve the envelope of the Gaussian pump (figure 5(c)), while the backscattered intensity (figure 5(f)) remains at negligible levels ($I_2(0) < 10 \text{ kW cm}^{-2}$).

Our results are summarized in figure 6(a), which shows the product of initial pump intensity $I_1(0)$ and self-focusing distance $z_c$ for input powers $< 30P_{cr}$. In the absence of SBS, the SF distances satisfy the Marburger formula (solid curve). With SBS, SF produces a nonlinear focus that barely varies with the pump power, as justified above. In contrast, when the pump is modulated in phase, the product $I_1(0) \times z_c$ saturates around 20 GW cm$^{-1}$, which remarkably agrees with the experimental data of [6] recalled by star symbols. This behavior can be explained by the pulse spatiotemporal breakup undergone by the multimode pump (see figure 5(b)). The SF distance then fulfills the relationship $z_c \simeq L_{SF} \times E_1(0)/E_1(L)$ for small $R$, such that $I_1(0) \times z_c \simeq 18 \text{ GW cm}^{-1}$. Pink triangles on the Marburger curve refer to amplitude-modulated pumps, for which SBS is completely suppressed. Figure 6(b) reports similar results computed for the infrared laser wavelength $\lambda_0 = 1064 \text{ nm}$ ($P_{cr} = 4.27 \text{ MW}$). We observe the same dynamics and excellent agreement with experimental data [6]. Pump collapse occurs whenever $P_1(0) > 3P_{cr}$ and the product $I_1(0) \times z_c$ saturates around the value of $I_1(0) \times L_{SF} \times E_1(0)/E_1(L) \simeq 80 \text{ GW cm}^{-1}$ in the case of phase modulation. These types of behavior are generic at any wavelength in transparent bulk materials. They were found to also hold for various values of mode numbers $m$ and modulation frequencies $\nu_m > 2 \text{ GHz}$ keeping $\Delta \nu$ in the range of a few hundreds of GHz (not shown).
In summary, we have numerically cleared up the interplay between stimulated Brillouin scattering and Kerr self-focusing driven by nanosecond pulses in fused silica. Particular attention was paid to power regimes allowing full self-focusing of the pump pulse. 3D numerical simulations emphasized the strong amplification of the Stokes intensity for phase-modulated pump lasers with bandwidths of the order of 100 GHz. To suppress SBS, we instead propose to exploit amplitude modulations with periods less than the Brillouin phonon lifetime.

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