Stimulated Raman Scattering of Rapidly Amplified Short Laser Pulses

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The theory of transient forward stimulated Raman scattering (FSRS) of rapidly amplified short laser pulses is put forth to complement the classical theory for FSRS of stationary pulses. Quantitative conditions for FSRS suppression are identified. In particular, it is shown quantitatively how the limitation imposed by pumped pulse FSRS on the output laser intensity in plasma-based ultrapowerful backward Raman amplifiers can be overcome through a selective detuning of the Stokes resonance.

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The classical theory for transient (near-)forward stimulated Raman scattering (FSRS) of laser pulses [1] has been worked out for arbitrary shape but stationary (nonevolving) pulses. However, the theory of FSRS of rapidly evolving laser pulses has not been worked out. Such a theory is necessary to describe evolving laser pulses in a variety of applications, including laser amplifiers, tightly focused laser pulses, self-focused pulses, and any case when the FSRS effect itself plays a role in evolving the parent pulse.

This Letter considers a short laser pulse rapidly amplified through Raman backscattering of a long pulse, and simultaneously experiencing FSRS. Remarkably, attractor solutions can be found not only to this rather generally posed problem but even for the case when the interactions are detuned off resonance. Thus we calculate precisely both the forward Raman leakage and its suppression in a detuning gradient.

This suppression enhances the prospect of achieving very high power laser pulses through fast backward Raman amplification (BRA) [2]. This scheme might enable the generation of laser energies and powers significantly higher than presently available through the most advanced chirped pulse amplifiers [3]. The proposed working medium is plasma, capable of tolerating ultrahigh laser intensities. Within times shorter than it takes for filamentation instabilities to develop, the transient backward stimulated Raman scattering (BSRS) of a laser pump in plasma is a fast enough amplification mechanism to reach nearly relativistic pumped pulse intensities, say 10^{17} W/cm^2, for $\lambda =$ 1-μm-wavelength radiation [2]. The nonfocused intensity would then be 10^5 times higher than currently available.

Traditional BRA devices were plagued by parasitic FSRS [4], which is faster than the BSRS in gases, liquids, and solids. In plasma, however, the BSRS is faster than the FSRS [5], which both alleviates and shifts the problem. A dangerous instability is then the BSRS of the pump to noise, as the pump traverses the plasma layer towards the seed pulse. This instability can be suppressed, however, by detuning the resonance appropriately, even as the desired amplification process persists with high efficiency due to nonlinear resonance broadening [6].

The problem of FSRS of the pumped pulse, however, still persists. The forward-to-backward Raman gain ratio is low in plasma, but the forward-scattered signal propagates together with the parent laser, and so has more distance to grow than does the backscattered signal (which quickly passes through a short parent pulse). This FSRS instability has been shown to impose the major theoretical limit on the peak intensity of output laser pulse in the suggestion of fast compression by BSRS [2,6].

What we now show and precisely calculate is how the parasitic FSRS of the rapidly evolving pumped laser pulse can be suppressed by employing a detuning gradient. The theoretical limit of the achievable laser output intensity is then raised, which is important for applications requiring very high power short pulses (see, e.g., [3,7]).

To begin, we first calculate the parasitic FSRS for the general resonant case. The resonant nonlinear dynamics, whether the coupling medium is a solid, liquid, gas, or plasma, is described by canonical equations for two coupled resonant three-wave interactions occurring in a broad range of phenomena [8,9],

$$\begin{aligned} a_t + c_a a_z &= -V_1 f b, \qquad f_t = V_1 a b^*, \\ b_t + c_b b_z &= V_1 a f^* - V_2 s l, \\ s_t + c_s s_z &= V_2 b l^*, \qquad l_t = V_2 b s^*. \end{aligned} \tag{1}$$

Here a, b, and s are amplitude envelopes of, respectively, (long) pump laser, (short) pumped pulse, and his first Stokes component; f and l are resonant material waves involved in the a-b and b-s interactions, respectively; V_1 and V_2 are three-wave coupling constants for these interactions, real for appropriately defined wave envelopes; subscripts t and z signify time and space derivatives; c_a, c_b, and c_s are group velocities of the pump, pumped pulse, and Stokes, respectively; and the group velocities of the material waves are neglected in comparison to those of lasers. For BRA, the pump and pumped lasers are counterpropagating, so that $c_a c_b < 0$, and the resonant material wave is of a relatively short wavelength ($|k_f| > |k_b|$). For FSRS of the pumped pulse, $c_s c_b > 0$, and the resonant material wave is of a relatively long wavelength ($|k_l| < |k_b|$). In new variables $\zeta = V_1(t - z/c_b)$, $\tau = V_1 z/c_b$, and notations $R \equiv V_2/V_1$, $R_{a;s} \equiv c_{a;s}/c_b \equiv 1 - r_{a;s}$, Eqs. (1) take the form

$$R_a a_\tau + r_a a_\zeta = -fb, \; f_\zeta = ab^*, \; b_\tau = af^* - Rsl, \tag{2}$$

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$$R_s s_\tau + r_s s_\zeta = Rbl^*, \qquad l_\zeta = Rbs^*. \tag{3}$$

As long as the Stokes influence on the pumped pulse can be neglected, the solution of Eqs. (2) is close to that for $R = 0$, which is known. During the linear stage of the backscattering instability [10], when the pump depletion can be neglected ($a \approx a_0 = \text{const}$), the originally small and narrow seed pulse is amplified and broadened. Its maximum increases with the peak growth rate for the monochromatic wave instability $a_0 V_1$ and moves with the speed $c_b/2$ (so that the distance between the maximum and the front of the pulse increases with the speed $c_b/2$). In the nonlinear stage, the amplification time increases (since a fixed pump acts relatively more slowly on larger signals), while the pump depletion scale decreases inversely proportional to the pumped pulse amplitude, and the pulse maximum moves with a super-luminous speed, approaching the front. An advanced nonlinear behavior is approximated well by a quasi-self-similar attractor solution, a so-called "π pulse," where the pump is fully depleted by the pumped pulse undergoing amplification and contraction [2,8]. During this stage the ζ scale is much smaller than the τ scale, and the term $|a_\tau| \ll |a_\zeta|$ in (2) can be neglected.

Consider now Eqs. (2) and (3) with $R \neq 0$. Assume, at first, that both the a_τ and s_τ terms can be neglected during a sufficiently advanced nonlinear stage. In the absence of the a_τ and s_τ terms, Eqs. (2) and (3) have integrals $|f|^2 + r_a|a|^2 = \text{const}_1$ and $|l|^2 - r_s|s|^2 = \text{const}_2$. For an intense enough pump ($r_a a_0^2 \gg f_0^2$) and FSRS seeded by the long-wavelength material wave noise ($r_s s_0^2 \ll l_0^2$), the integration constants are $\text{const}_1 \approx r_a a_0^2$ and $\text{const}_2 \approx l_0^2$. For $r_s > 0$ and $r_a > 0$, solutions of Eqs. (2) and (3) can be approximated by the following special solution:

$$a = a_0 \cos(\tfrac{u}{2}), \quad f = \sqrt{r_a}\, a_0 \sin(\tfrac{u}{2}), \quad b = \sqrt{r_a}\, \tfrac{u_\zeta}{2}, \tag{4}$$

$$l = l_0 \cosh(Ku/2), \qquad s = l_0 \sinh(Ku/2)/\sqrt{r_s}, \tag{5}$$

$$u_{\zeta\tau} = a_0^2 \sin u - K l_0^2 \sinh(Ku)/r_a, \quad K \equiv R\sqrt{r_a/r_s}\,. \tag{6}$$

Equation (6) for the integrated amplitude of the pumped pulse $u\sqrt{r_a}/2 = \int_{-\infty}^{\zeta} d\zeta_1\, b$ allows the self-similar substitution $u = u(\xi)$, $\xi = 2a_0\sqrt{\zeta\tau}$ which reduces it to the ordinary differential equation (ODE)

$$u_{\xi\xi} + \frac{u_\xi}{\xi} = \sin u - Q\sinh(Ku), \qquad Q \equiv \frac{K l_0^2}{a_0^2 r_a}. \tag{7}$$

For $Q = 0$, Eq. (7) reduces to the sine-Gordon equation, which has the classical π-pulse solution that depends on only a single parameter $u(+0) \equiv \epsilon$ [2,8]. Of interest here is the case of a small initial integrated amplitude of the seed pulse $\epsilon \ll 1$. Then, u reaches its maximum value close to 2π in the first zero of b, following the leading b spike in the π pulse wave train, and afterwards oscillates with a decreasing sweep around the stable stagnation point $u = \pi$ reached at $\xi \to \infty$.

For $Q \neq 0$, as seen from the modified sine-Gordon equation (6),(7), the π-pulse regime of the BRA is not affected noticeably by the FSRS as long as the condition $Q\sinh(2\pi K) < 1$ is satisfied. For $K \gg 1$ this implies an exponentially small value of Q, i.e., an exponentially small noise-to-pump intensity ratio. On other hand, the BRA is not completely suppressed by the FSRS but rather persists under a much more relaxed condition $QK < 1$, as seen immediately from Eq. (7) linearized over u. Thus, there is a broad range of parameter values where the BRA persists, but clearly affected by the FSRS.

The efficiency of BRA remains high as long as the leading b-spike maximum, where $u \approx \pi$, is not strongly affected, i.e., at $Q\sinh(\pi K) < 1$. The same condition implies that the stable stagnation point u_∞ [$\sin u_\infty = Q\sinh \times (K u_\infty)$], to which u tends at $\xi \to \infty$, remains close to π, so that the pump is nearly completely depleted. For larger noise, such that $Q\sinh(\pi K) > 1$, the pump depletion $1 - a_\infty^2/a_0^2 = \sin^2(u_\infty/2)$ decreases. Say $Q\sinh(\pi K/2) = 1$ corresponds to $u_\infty = \pi/2$, so that the depletion is 50%. A numerical calculation of FSRS-affected BRA self-similar regimes supports these assertions, as shown in Fig. 1.

The relative intensity of the output Stokes pulse is $s_\infty^2/a_0^2 = Q r_a \sinh^2(K u_\infty/2)/K r_s \approx \sin u_\infty \sqrt{r_a/r_s}/2R$, for $K u_\infty \gg 1$. It increases with the noise for $l_0^2 < a_0^2 r_a / 2K\sinh^2(K\pi/4)$, corresponding to $\pi > u_\infty > \pi/2$. For an even higher noise, such that $u_\infty < \pi/2$, yet $K u_\infty \gg 1$, the Stokes intensity s_∞^2 decreases with the noise intensity l_0^2, because of the BRA suppression.

The numerical solution of the initial value problem for Eqs. (2) and (3) indicates that their advanced solutions are indeed well approximated by those of Eq. (6), which in turn tend to the self-similar solutions given by the ODE (7), $b/\tau a_0^2\sqrt{r_a} \to u_\xi/\xi$. It indicates that modified self-similar solutions are attractors for solutions of Eqs. (2) and (3).

The tolerable level of noise (seeding FSRS) can be increased substantially by using an appropriately detuned BRA scheme. To include frequency detunings, Eqs. (1), for material waves should be modified as follows:

$$f_t + \imath\Omega_f f = V_1 a b^*, \qquad l_t + \imath\Omega_l l = V_2 b s^*, \tag{8}$$

where $\Omega_f \equiv \omega_f + \omega_b - \omega_a$, $\Omega_l \equiv \omega_l + \omega_s - \omega_b$, and $\omega_{a,b,s,f,l}$ are respective wave frequencies. Detunings Ω_f and Ω_l can be varied independently by using two different detuning mechanisms, the pump frequency chirp and the Raman frequency variation. For a short enough pumped pulse, detunings in the pulse location z are well-defined functions of z. Provided these functions are smooth enough, they can be linearized over z near exact resonance points. Let these points be located at $z = 0$, then $\Omega_f = \Omega_f' z \equiv q_f a_0^2 V_1 \tau$ and $\Omega_l = \Omega_l' z \equiv q_l a_0^2 V_1 \tau$, where q_f and q_l are dimensionless detuning gradients. Changing variables t and z for ζ and τ, one can note that, in the regime where a_τ and s_τ terms are negligible, the above similarity survives even with the detunings. The self-similar substitution

$$b = a_0^2 \tau \sqrt{r_a}\, B(\eta), \quad \eta = a_0^2 \zeta\tau = \xi^2/4, \quad a = a_0 A,$$
$$f = a_0\sqrt{r_a}\, F, \quad l = l_0 L, \quad s = l_0 S/\sqrt{r_s} \tag{9}$$

leads then to the following ODE set:

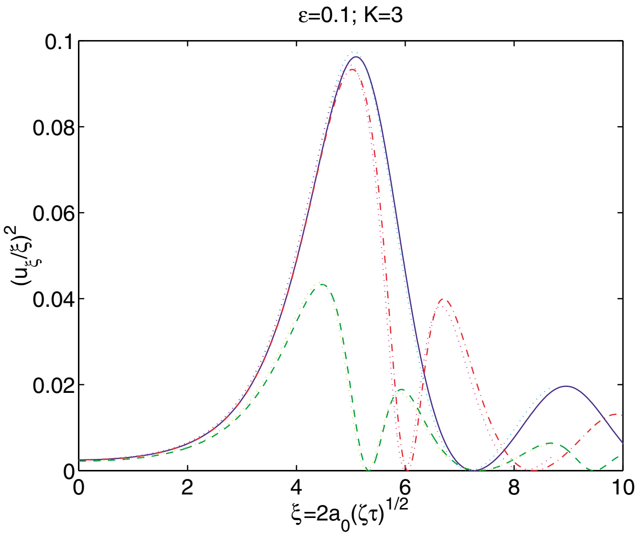

FIG. 1 (color). The rescaled pumped pulse intensity $(u_\xi/\xi)^2$ for $\epsilon = 0.1$, $K = 3$, and different values of effective noise intensity Q, namely, $Q = 0$, π-pulse solution—solid line; $Q = 1/\sinh(\pi K)$—dash-dotted line; $Q = 1/\sinh(\pi K/2)$—dashed line. The rescaled pumped pulse intensity $|b/\tau a_0^2\sqrt{r_a}|^2$ computed from the respective initial value problem with the same ϵ, K, and Q is shown by dotted lines.

$$(\eta B)_\eta = AF^* - QSL, \quad A_\eta = -BF, \quad S_\eta = KBL^*,$$
$$F_\eta + iq_f F = AB^*, \quad L_\eta + iq_l L = KBS^*. \qquad (10)$$

For $q_f = q_l = 0$, this set has real solutions given by the modified sine-Gordon equation (7). The BRA detuning gradient $q_f \sim 0.25$ prevents a premature pump backscattering by noise, while the useful amplification process persists with a high efficiency. This nonlinear filtering effect was in fact shown in [6] under conditions when the FSRS is negligible. What we show now is that even a strongly seeded FSRS can be suppressed by means of the Stokes detuning gradient q_l. The BRA efficiency approaches then the limit where FSRS is absent, as shown in Fig. 2. An estimate for the FSRS gain can be obtained by the WKB method, assuming that the parent pulse B is unaffected by the FSRS. Then, for $L \propto S^* \propto e^\sigma$, one gets in the zeroth-order WKB approximation $\sigma_\eta = \sqrt{K^2|B|^2 - q_l^2/4} - iq_l/2$. For $q_l = q_f = 0$, when $B = u_\eta/2$, it gives $\sigma = Ku/2$ which agrees with the above formulas for the exactly resonant case. In slightly detuned regimes, a noticeable FSRS suppression should occur at $q_l \sim KB_*$, where $B_* \equiv \max_\eta |B| = |B(\eta_*)|$, and nearly complete FSRS suppression should occur at $q_l \sim 2KB_*$. The numerical solution of ODE (10), presented in Fig. 2, confirms these crude estimates. The figure also shows that the detuned self-similar regimes are attractors for solutions of the initial value problem.

For the above self-similar solutions, where both a and s depend just on the product $\tau\zeta$, the conditions that the τ derivatives be small in equations for the pump and Stokes waves, i.e., $|R_a a_\tau| < r_a|a_\zeta|$ and $R_s|s_\tau| < r_s|s_\zeta|$, can be written as $\tau/\sqrt{\eta} > \sqrt{|R_a/r_a|}/a_0 \equiv \tau_a$ and $\tau/\sqrt{\eta} > \sqrt{R_s/r_s}/a_0 \equiv \tau_s$, respectively. Under usual conditions when $R_s \approx 1 \gg r_s$, while $R_a \approx -1 \Leftrightarrow r_a \approx 2$, there is a broad intermediate range $\tau_a < \tau/\sqrt{\eta} < \tau_s$, where the Stokes convective term is still negligible, while the pump is already quasistatic. As long as the Stokes influence on the pumped pulse can be neglected, the Stokes evolution is described by linear nonstationary equations

$$s_\tau = Rbl^*, \qquad l_\zeta + \imath(\Omega_l/V_1)l = Rbs^*. \qquad (11)$$

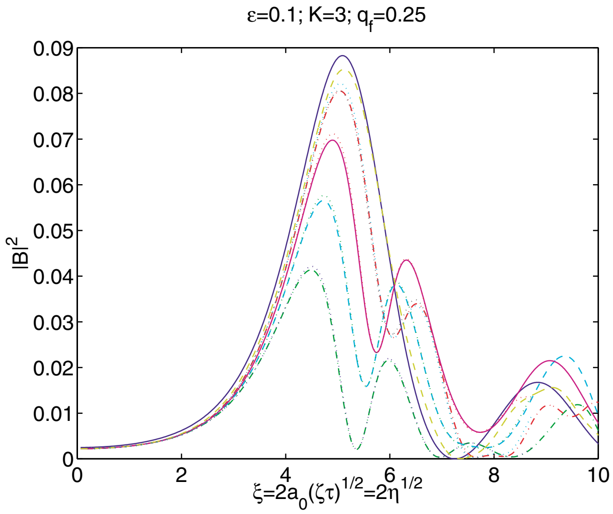

FIG. 2 (color). The rescaled pumped pulse intensity $|B|^2$ for $\epsilon = 0.1$, $K = 3$ and the BRA detuning gradient $q_f = 0.25$; the upper solid line corresponds to zero noise $Q = 0$; all other lines correspond to $Q = 1/\sinh(\pi K/2)$ and different values of the FSRS detuning gradient q_l, namely, $q_l = 0$—the lower dash-dotted line, $q_l = 0.5$—the lower dashed line, $q_l = 0.75$—the lower solid line, $q_l = 1$—the upper dash-dotted line, and $q_l = 1.5$—the upper dashed line. The rescaled intensity $|b/\tau a_0^2\sqrt{r_a}|^2$ computed from the respective initial value problem is shown by dotted lines.

The potentially dangerous regimes where Stokes is greatly amplified can be analyzed by the WKB method. For $s^* \propto l \propto e^\sigma$, the zero-order WKB approximation leads to the equation $\sigma_\tau(\sigma_\zeta + \imath\Omega_l/V_1) = |Rb|^2$. For b from Eq. (9), this equation, with $\Omega_l = 0$, i.e., without the detuning, has the self-similar solution $\sigma = a_0\tau R\sqrt{r_a}\,\psi(\eta)$, $(\eta\psi_\eta + \psi)\psi_\eta = |B|^2$, that gives the Stokes growth rate at each point η. To find the detuning needed to reduce substantially the Stokes growth, consider formally $\Omega_l = \Omega_{lm} = Kq_{lm}V_1 a_0^2\tau^2/\tau_s$. Then, the self-similar substitution $\sigma = a_0\tau R\sqrt{r_a}\,\psi(\eta)$ survives, leading to the following equation for ψ:

$$(\eta\psi_\eta + \psi)(\psi_\eta + \imath q_{lm}) = |B|^2. \qquad (12)$$

For $q_{lm} = 0$, the maximum of ψ_η can be crudely evaluated as $B_*/\sqrt{\eta_*}$. The detuning $q_{lm} \sim B_*/\sqrt{\eta_*}$ should reduce noticeably the FSRS effective growth rate $\Re\psi$. The numerical solution of (12), presented in Fig. 3, confirms this crude estimate.

The detunings needed for the FSRS suppression in the convectionless and convective regimes are $\Omega_{lm} \sim KB_*V_1a_0^2\tau^2/\tau_s\sqrt{\eta_*}$ and $\Omega_l \sim 2KB_*V_1a_0^2\tau$, respectively. The detuning gradients are related by $\Omega_{lm}^l \sim \Omega_l^l\tau/\tau_s\sqrt{\eta_*}$.

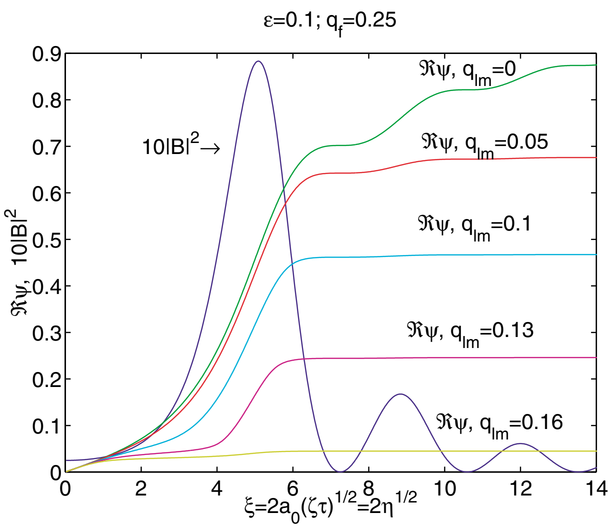

FIG. 3 (color). The effective growth rate $\Re\psi$ of convectionless FSRS in the BRA regime with initial integrated seed amplitude $\epsilon = 0.1$ and BRA detuning gradient $q_f = 0.25$, for several values of the modified FSRS detuning parameter q_{lm}.

Up to $\tau \sim \tau_s\sqrt{\eta_*}$, when the transition between the regimes in the location of the pumped pulse maximum should occur, Ω'_{lm} is smaller than Ω'_l, i.e., the detuning gradient suppressing FSRS in convective regimes is sufficient for the FSRS suppression during the earlier BRA stages as well.

Consider now the application of the above theory to powerful plasma-based BRA. In a cold plasma, $\omega_l = \omega_f = \omega_p \equiv \sqrt{4\pi n_e e^2/m_e}$, where m_e, e, and n_e are electron mass, charge, and concentration, respectively. For the laser frequency ω substantially exceeding the plasma frequency, $\omega_p \ll \omega_s \approx \omega_b \approx \omega_a$, the laser group velocities $v \approx c(1 - \omega_p^2/2\omega^2)$ are close to the speed of light c, and $R_a \approx -1$, $r_a \approx 2$, $R_s \approx 1$, while $r_s \approx \omega_p^3/\omega^3 \ll 1$. The FSRS to BSRS coupling constant ratio is $R \approx \omega_p/2\omega \ll 1$, so that $K \equiv R\sqrt{r_a/r_s} \approx \sqrt{\omega/2\omega_p} > 1$. Then, $Q = Kl_0^2/2a_0^2 = K^3 W_l/W_a$, where W_a and W_l are initial energy densities of the pump and long Langmuir wave, respectively. Note that, according to the above theory, the FSRS is sufficiently suppressed by the convection for $W_l \ll W_a/K^3 \sinh(\pi K)$, but otherwise an extra suppression mechanism, like detuning, is needed.

For the undercritical plasma considered here, it is convenient to specify quantities a, b, and s as space-time envelopes of the respective electron quiver velocities measured in the units of the light speed c. The normalization $I_a = cW_a = \pi c(m_e c^2/e)^2|a|^2/\lambda^2 = 2.736 \times 10^{18}|a|^2/\lambda^2[\mu\text{m}]$ W/cm^2, where I_a and λ are the laser intensity and wavelength, and similar normalizations for b and s correspond to the BRA coupling constant $V_1 = \sqrt{\omega\omega_p/2}$, both for linearly and circularly polarized lasers. The envelopes f and l of respective Langmuir wave electric fields E_f and E_l are then specified by $E_f e/V_1 m_e c = fe^{\imath(k_f z - \omega_p t)}$ + c.c. and similar for l.

The slowly varying envelope approximation for the short-wavelength Langmuir wave is valid as long as $|f_t| < \omega_p|f| \Leftrightarrow V_1|b| < \omega_p \Leftrightarrow \tau a_0^2 B_*\sqrt{\omega/\omega_p} < 1$. The similar requirement to the long-wavelength Langmuir wave in the convective regime is stricter: $|l_t| < \omega_p|l| \Leftrightarrow \Omega_l < \omega_p \Leftrightarrow \tau a_0^2 B_*\omega/\omega_p < 1$. This may be satisfied in the domain $\tau > \tau_s\sqrt{\eta_*} = \sqrt{\eta_*}/a_0\sqrt{r_s}$, where the convective regime occurs, for $a_0 < (\omega_p/\omega)^{5/2}/B_*\sqrt{\eta_*} \sim 1.3(\omega_p/\omega)^{5/2}$. For a reasonable frequency ratio, like $\omega/\omega_p \lesssim 10$, the latter condition does not differ much from the condition that the short-wavelength Langmuir wave is not broken, $4a_0 < (\omega_p/\omega)^{3/2}$ [2], which is assumed anyway in regimes considered here. Conditions that the pumped pulse dispersion and relativistic electron nonlinearity are negligible in the beginning of the convective regime are $a_0 < 3(\omega_p/\omega)^2$ and $a_0 < 3(\omega_p/\omega)^3$, respectively.

Anti-Stokes excitation is described by the equation $\bar{s}_t + c_b(1 + r_s)\bar{s}_z + \imath\omega_p r_s\bar{s} = -V_2 bl$. In the convective regime, where the approximation $\bar{s}_t + c_b\bar{s}_z = 0$ is valid, anti-Stokes is decoupled from Stokes resonance and suppressed, since $c|\bar{s}_z| < \omega_p|\bar{s}|$ as long as the slowly varying envelope approximation is applicable. The numerical solution of equations including anti-Stokes confirms that anti-Stokes is indeed small in the whole applicability range of our equations neglecting relativistic nonlinearity. This may be an experimentally verifiable signature of the new regimes, since for longer pulses, anti-Stokes excitation occurs at much smaller intensities.

In summary, apart from the broad applicability of our results within the general theory of three-wave interactions, an important application is FSRS suppression in powerful plasma-based BRA. By operating in a Raman detuning gradient, huge FSRS-seeding noise might then be tolerated in such devices without being amplified significantly, while the useful amplification persists with high efficiency.

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