Raman Forward Scattering of High-Intensity Chirped Laser Pulses

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Abstract. Raman forward scattering of a high-intensity, short-duration, frequency-chirped laser pulse propagating in an underdense plasma is examined. The growth of the direct forward scattered light is calculated for a laser pulse with a linear frequency chirp in various spatiotemporal regimes. This includes a previously undescribed regime of strongly-coupled four-wave nonresonant interaction, which is important for relativistic laser intensities. In all regimes of forward scattering, it is shown that the growth rate increases (decreases) for positive (negative) frequency chirp. The effect of chirp on the growth rate is relatively minor, i.e., a few percent chirp yields few percent changes in the growth rates. Relation of these results to recent experiments is discussed.

1. INTRODUCTION

High-intensity short-pulse laser-plasma interactions are of much current interest because of their application to laser-plasma accelerators [1], laser-plasma-based harmonic generation [2], x-ray lasers [3], and laser-driven inertial confinement fusion schemes [4]. A basic phenomenon in laser-plasma interactions is Raman scattering. The Raman scattering instability [5] is the resonant decay in a plasma of incident light, with frequency and wave number (ω_0, k_0) , into a plasma wave (ω, k) and scattered light $(\omega_0 \pm \omega, k_0 \pm k)$. Physically, the Raman instability occurs due to the beating of the incident and scattered light, producing a ponderomotive force which generates a plasma density modulation at or near the plasma frequency ω_p , where $\omega_p = (4\pi e^2 n_0 / m)^{1/2}$ with n_0 the equilibrium electron plasma density. The plasma density (index of refraction) modulation causes modulation of the incident laser pulse, resulting in additional scattering, thereby producing an instability. The transmission of laser light through a plasma, and the coupling of the laser energy to a plasma, can be greatly affected by Raman scattering, which consequently can have a large impact on various applications.

For example, Raman scattering in the forward direction (i.e., the scattered light is co-propagating with the incident light) can be used to drive the self-modulated laser wakefield accelerator (for a review see Ref. [1]), in which a long (compared to the plasma wavelength) laser pulse becomes modulated and produces a large amplitude plasma wave with phase velocity near the speed of light $v_\phi = \omega/k \simeq c$. This plasma wave, with 10-100 GeV/m accelerating gradients having been demonstrated using present laser technology, can be used to accelerate charged particles to high energies [6, 7, 8, 9]. In laser fusion applications, such as the fast-ignitor [4], the excitation of Raman instabilities can yield poor coupling of the laser to the energetic electrons. The use of

finite-bandwidth laser pulses has been considered for enhancement or suppression of Raman instabilities [10, 11], and therefore as a means to control Raman scattering in these applications.

Several ultra-intense laser facilities around the world have been investigating the effect of frequency-chirped (i.e., frequency correlated to longitudinal position within the pulse) laser pulses propagating in an underdense plasma [12, 13, 14]. Experimental evidence by Faure *et al.* [12] has shown that the growth of the Raman instabilities is independent of the frequency chirp. Other experiments by Yau *et al.* [13] have reported enhanced efficiency of the Raman forward scattering instability for positively-chirped laser pulses, and recent experiments by Leemans *et al.* [14] reported frequency chirp induced asymmetries in the self-modulated laser wakefield electron yield. In addition, two-dimensional particle-in-cell (PIC) computer simulations presented in Ref. [15] claim enhancement of Raman forward scattering instabilities for positively-chirped laser pulses, however, these simulations assumed a large bandwidth (20%), an order of magnitude beyond that used in present-day experiments [12, 13, 14].

In this paper, we analyze the Raman forward scattering (RFS) of a short frequency-chirped laser pulse of relativistic-intensity propagating in an underdense plasma, and calculate the effect of a correlated frequency chirp on the growth of the Raman instability using the coupled relativistic Maxwell-fluid equations. In Sec. 2, we review the basic Maxwell-fluid equations for the study of laser propagation in an underdense plasma used in the instability analysis. In Sec. 3, the spatiotemporal growth of the plasma wave generated by RFS is calculated for a laser pulse with a linear frequency chirp in various spatiotemporal regimes. This includes a previously undescribed strongly-coupled four-wave nonresonant regime, which is important for relativistic laser intensities. It is shown that the growth rate increases (decreases) for positive (negative) chirp in all regimes of the RFS instability. The RFS growth rates are summarized in Sec. 3.6. In Sec. 3.7 we examine the asymmetry between plasma wave generation using a laser pulse with positive and negative frequency chirp. It is shown that the effect of chirp on the growth rate is relatively minor, i.e., a few percent chirp yields few percent changes in the growth rates. Section 4 presents a summary of the results, and discusses their relation to recent experiments.

2. BASIC FORMULATION AND ASSUMPTIONS

A one-dimensional (1D) model of the laser-plasma interaction is considered. The 1D model will be valid provided $r_s \gg \lambda_p$, where r_s is the laser spot size and λ_p is the plasma wavelength. The plasma wavelength is defined as $\lambda_p^2 = \pi/(n_0 r_e)$, where $r_e = e^2/(mc^2)$ is the classical electron radius. The 1D fields associated with the pump laser, scattered light, and plasma response can be described by the transverse vector and the scalar potentials. In the Coulomb gauge, the Maxwell equations for the fields can be expressed

in 1D as

$$\left[\frac{\partial^2}{\partial z^2} - \frac{\partial^2}{\partial (ct)^2} \right] a = k_p^2 \frac{n}{n_0} \frac{a}{\gamma}, \quad (1)$$

$$\frac{\partial^2 \phi}{\partial z^2} = k_p^2 \left(\frac{n}{n_0} - 1 \right), \quad (2)$$

where $k_p = 2\pi/\lambda_p$, and a and ϕ are the transverse vector and scalar potentials respectively normalized to mc^2/e . Note that in the 1D approximation, conservation of transverse canonical momentum yields $\gamma\beta_\perp = a$ for an initially quiescent plasma, where $\gamma = (1 - \beta_z^2 - \beta_\perp^2)^{-1/2}$ is the relativistic Lorentz factor, and β_z and $\beta_\perp$ are the electron longitudinal and transverse fluid velocities respectively normalized to the speed of light.

A cold-fluid model of the neutral plasma is assumed. Thermal effects may be ignored when the quiver velocity is much greater than the electron thermal velocity and the thermal energy spread is sufficiently small such that electron trapping in the plasma does not take place. The ions are also assumed to be stationary, which is typically the case for short-pulse (< 1 ps) laser interactions in underdense plasmas. The cold fluid equations can be expressed in 1D as

$$\frac{\partial}{\partial t}(\gamma\beta_z) = \frac{\partial}{\partial z}(\phi - \gamma), \quad (3)$$

$$\frac{\partial n}{\partial t} + \frac{\partial}{\partial z}(n\beta_z) = 0, \quad (4)$$

where n is the plasma number density.

It is convenient to work in the comoving variable $\zeta = z - ct$ and transform from the variables (z, t) to $(\zeta = z - ct, \tau = ct)$. We will assume that the head of the right-going laser pulse is initially at $\zeta = 0$ and the body of the laser pulse extends into the region $\zeta \leq 0$, while the plasma is unperturbed in the region $\zeta > 0$. To study the growth of Raman instabilities, consider a density perturbation $\delta n = n/n_0 - 1$, which results from the scattering of a large-amplitude pump laser pulse a_{pump} into daughter waves a_{scat} , such that $|a_{\text{pump}}| \gg |a_{\text{scat}}|$. Linearizing about the perturbations δn and a_{scat} , Eqs. (1) - (4) can be combined to yield, in comoving variables (ζ, τ) ,

$$\left[\left(\frac{\partial}{\partial \tau} - \frac{\partial}{\partial \zeta} \right)^2 + \frac{k_p^2}{\gamma_{\perp 0}} \right] \delta n = \frac{1}{\gamma_{\perp 0}^2} \frac{\partial^2}{\partial \zeta^2} (a_{\text{pump}} a_{\text{scat}}), \quad (5)$$

and

$$\left[2 \frac{\partial^2}{\partial \zeta \partial \tau} - \frac{\partial^2}{\partial \tau^2} - \frac{k_p^2}{\gamma_{\perp 0}} \right] a_{\text{scat}} = \frac{k_p^2}{\gamma_{\perp 0}} a_{\text{pump}} \left[\delta n - \frac{1}{\gamma_{\perp 0}^2} (a_{\text{pump}} a_{\text{scat}}) \right], \quad (6)$$

where $\gamma_{\perp 0}^2 = 1 + a_{\text{pump}}^2$.

We model the pump and scattered laser pulse normalized transverse vector potentials (linear polarization is assumed) as

$$a = a_{\text{pump}} + a_{\text{scat}} = \frac{a_0}{2} e^{i\varphi_0} + \sum_{\pm} \frac{a_{\pm}}{2} e^{i\varphi_{\pm}} + \text{c.c.}, \quad (7)$$

where a_0 is the pump amplitude, and $a_{\pm}$ are the slowly-varying envelopes of the Stokes (down-shifted) and anti-Stokes (up-shifted) scattered light waves. We will assume $|a_0| \gg |a_{\pm}|$ and a_0 is a nonevolving envelope (i.e., pump depletion effects are neglected). To lowest-order, $a^2 \simeq a_{\text{pump}}^2 = a_0^2/2$. The temporal and spatial derivatives of the phase determine the local values of the pulse frequency and wave number: $\omega_i = -\partial\varphi_i/\partial t$ and $k_i = \partial\varphi_i/\partial z$. The plasma density perturbation is modeled as $\delta n = (\hat{n}/2)\exp(i\varphi_p) + \text{c.c.}$, where $\hat{n}$ is the slowly-varying envelope of the plasma density perturbation and $\varphi_p = kz - \omega t$. The resonance condition for 1D Raman scattering requires $\varphi_{\pm} = \varphi_0 \pm \varphi_p$.

The effect of a finite-bandwidth on parametric instabilities, such as the Raman instability, has been extensively studied [10, 11] for the case of an uncorrelated, or random, frequency bandwidth. In this work, we examine the effect of a correlated frequency chirp on the Raman instability growth rates. Note that, if we consider a linear frequency chirp $\varphi_0 = k_0\zeta + \varepsilon\zeta^2$ on a pump laser pulse with constant root-mean square (rms) frequency bandwidth σ_k and a Gaussian intensity distribution $a_0 = \hat{a}_0 \exp[-\zeta^2/(4\sigma_z^2)]$, where σ_z is the rms pulse length of the laser intensity, then the linear chirp will be given by $\varepsilon = \{[\sigma_k/(2\sigma_z)]^2 - (2\sigma_z)^{-4}\}^{1/2}$. Therefore the full-width-half-maximum (FWHM) relative chirp over the Gaussian pulse is $\Delta k_e/k_0 = 2\varepsilon L/k_0 = \sqrt{2\ln 2}[(k_0\sigma_0)^{-2} - (k_0\sigma_z)^{-2}]^{1/2}$, where σ_0 is the Fourier transform-limited pulse length. To isolate the effect of a correlated frequency chirp we will consider laser pulses such that $k_p\sigma_z \gg 1$, where σ_z is the root-mean square (rms) pulse length of the laser intensity, and the laser pulse intensity is approximately uniform within the pulse. We will also focus our analysis on analytic solutions describing the Raman instability with the condition $k_p^2/(k_0^2\gamma_{\perp 0}) \ll 1$, i.e., laser propagation in an underdense plasma with group velocity of the laser approximately the speed of light.

3. RAMAN FORWARD SCATTERING

In RFS, the scattered waves, which are referred to as the Stokes (down-shifted, $k_0 - k$) and anti-Stokes (up-shifted, $k_0 + k$) waves, propagate in the same direction as the pump laser pulse $k_{\pm} \sim k_0 \gg k \sim k_p$. To solve Eqs. (5) and (6) for the growth of the RFS instability, it is convenient to define $\chi = (a_0^*a_+ + a_0a_-^*)/(2\gamma_{\perp 0}) - \gamma_{\perp 0}\hat{n}$. In the quasi-static approximation [16], χ is simply the amplitude of the plasma wave potential perturbation $\delta\phi = \phi - \phi_0 = (\chi/2)\exp(kz - \omega t) + \text{c.c.}$, where $\phi_0 = \gamma_{\perp 0} - 1$ is the quasi-static equilibrium potential. Using the eikonal (slowly-varying amplitude) approximation $|\partial_{\tau}\chi| \ll |k\chi|$ and $|\partial_{\tau}a_{\pm}| \ll |ka_{\pm}|$, Eq. (5) reduces to

$$\left(\frac{\partial^2}{\partial\zeta^2} + 2ik\frac{\partial}{\partial\zeta}\right)\chi = \frac{k^2}{2\gamma_{\perp 0}}[a_0^*a_+ + a_0a_-^*], \quad (8)$$

where $k^2 = k_p^2/\gamma_{\perp 0}$ is the relativistic plasma wave number. With the eikonal approximation $|\partial_{\zeta}a_{\pm}| \ll |k_{\pm}a_{\pm}|$, the evolution equations for the daughter waves Eq. (6) reduces

to

$$\left[2i \left(\frac{\partial \varphi_+}{\partial \zeta} \right) \frac{\partial}{\partial \tau} + D_+ \right] a_+ = -\frac{k_p^2}{2\gamma_{\perp 0}^2} a_0 \chi, \quad (9)$$

$$\left[-2i \left(\frac{\partial \varphi_-}{\partial \zeta} \right) \frac{\partial}{\partial \tau} + D_- \right] a_-^* = -\frac{k_p^2}{2\gamma_{\perp 0}^2} a_0^* \chi, \quad (10)$$

where $D_{\pm} = \omega_{\pm}^2/c^2 - k_{\pm}^2 - k_p^2/\gamma_{\perp 0}$ is the dispersion relation for each daughter wave. If the Stokes wave is assumed to be resonant $D_- = 0$, then $|D_+/k_0^2| \simeq 2k_p^4/(\gamma_{\perp 0}^2 k_0^4) \ll 1$.

For definiteness, in this work we will consider a pump laser pulse with a flat-top distribution such that $a_0(\zeta) = a_0$ for $\zeta \in [-L, 0]$ (i.e., the head of the pulse is located at $\zeta = 0$ and the tail of the pulse at $\zeta = -L$), and a linear chirp on the pump laser pulse $\varphi_0 = k_0 \zeta + (\Delta k_e/2) \zeta (1 + \zeta/L)$. The local wave number is $k_{0l} = \partial_{\zeta} \varphi_0 = k_0 + \Delta k_e (\zeta/L + 1/2)$, such that k_0 is the central wave number and $\Delta_e \equiv \Delta k_e/k_0$ is the relative chirp over the FWHM pump laser pulse length. By assuming this form of the pump laser pulse, we are neglecting pump dispersion effects. This is justified since the growth length of the Raman instabilities is typically much shorter than the characteristic length for dispersive broadening $Z_D = k_0^3 L^2 / (2k_p^2)$ [17], i.e., $k_p Z_D = (k_p L)^2 (k_0/k_p)^3 / 2 \gg \sqrt{8} \gamma_{\perp 0}^2 (k_0/k_p) / |a_0|$ for $a_0 \sim 1$, $k_0/k_p \gg 1$, and $k_p L \sim 1$ (considering the rate at which a single modulation of length $\sim \lambda_p$ disperses).

Several regimes of RFS can be identified [18, 19, 20], and, as the instability grows, it passes through these various regimes depending on the value of $|\zeta|/\tau$ and the intensity of the incident laser pulse a_0 . Past analytical analysis on Raman instabilities [18, 19, 20] has primarily focused on nonrelativistic laser-plasma interactions where $a_0 \ll 1$. In this work we perform a relativistic analysis of the linear RFS spatiotemporal growth rates including a correlated frequency chirp. We also show that, for relativistic intensities $a_0 \gtrsim 1$, the RFS instability can enter a strongly-coupled regime where the growth rate of the instability becomes larger than the plasma frequency.

3.1. Four-wave resonant regime

Consider the four-wave resonant interaction where both the Stokes and anti-Stokes modes $k_0 \pm k_p$ are approximately resonant, and we will assume $D_+ \simeq 0$ and $D_- = 0$. We will also assume that we are in the weakly-coupled regime such that $|\partial_{\zeta} \chi| \ll |k \chi|$. Combining the envelope equations for plasma wave and scattered electromagnetic waves Eqs. (8) - (10) yields

$$\frac{\partial^2 \chi}{\partial \zeta \partial \tau} = \frac{k_p^4 a_0^* a_0}{16 \gamma_{\perp 0}^4 k} \left[\left(\frac{\partial \varphi_+}{\partial \zeta} \right)^{-1} - \left(\frac{\partial \varphi_-}{\partial \zeta} \right)^{-1} \right] \chi \quad (11)$$

$$\simeq -\frac{k_p^4 |a_0|^2}{8 \gamma_{\perp 0}^4} \left(\frac{\partial \varphi_0}{\partial \zeta} \right)^{-2} \chi = -\Gamma_{4\text{loc}}^2 \chi, \quad (12)$$

where

$$\Gamma_{4\text{loc}}(\zeta) = \frac{k_p^2 |a_0|}{\sqrt{8} \gamma_{\perp 0}^2 k_{1\text{loc}}} \quad (13)$$

is the relativistic growth rate for the four-wave resonant RFS instability [21] at the local wave number of the pump laser $k_{\text{loc}}(\zeta) = \partial_\zeta \phi_0$. We will assume the initial condition $\partial_\zeta \chi(\tau = 0, \zeta) = 0$, i.e., the amplitude of the initial seed plasma wave perturbation is constant throughout the pump laser pulse. Consider a source of noise at the head of the pulse $\chi(\tau, \zeta = 0) = \chi_0 H(\tau)$, where H is the Heaviside step function, i.e., the noise source at the front of the pulse is constant since the laser pulse is moving into fresh unperturbed plasma. The solution to Eq. (12) for the amplitude of the plasma wave potential inside the laser pulse (for $-L \leq \zeta \leq 0$) is

$$\chi(\tau, \zeta) = \chi_0 H(\tau) I_0 \left[2\Gamma_{\text{4eff}} \sqrt{\tau|\zeta|} \right], \quad (14)$$

where I_0 is the modified Bessel function of zeroth-order. Asymptotically $k_p^2 |\zeta| \tau \gg 1$, the amplitude of the plasma wave grows exponentially. The effective growth rate of the plasma wave due to the four-wave resonant RFS instability is a function of position within the pump laser pulse:

$$\Gamma_{\text{4eff}}(\zeta) = \Gamma_{\text{4loc}} \left[1 - \Delta_e \frac{|\zeta|}{L} \left(1 + \frac{\Delta_e}{2} \right)^{-1} \right]^{1/2}. \quad (15)$$

For a *positive* chirp (i.e., $\Delta_e < 0$, with red wavelengths at the head and blue wavelengths at the tail of the laser pulse), the RFS growth rate Eq. (15) is *greater* than the local growth rate throughout the laser pulse. Note that for the unchirped case, $\Delta_e = 0$, the effective growth rate reduces to $\Gamma_{\text{4eff}} = \Gamma_4 = k_p^2 |a_0| / (\sqrt{8} \gamma_{\perp 0}^2 k_0)$, the usual nonlinear growth rate for the four-wave resonant RFS instability [21]. For example, at the center of the pulse, $\Gamma_{\text{4eff}}(\zeta = -L/2) = \Gamma_4 (1 + \Delta_e/2)^{-1/2}$, and the growth rate is changed by only 2.4% due to a 10% chirp over the pump laser pulse ($\Delta_e = 0.1$). For $|\Delta_e| \ll 1$, $\Gamma_{\text{4eff}} \simeq \Gamma_4 [1 - (\Delta_e/2)(1 - |\zeta|/L)]$.

In deriving the four-wave resonant RFS growth rate it was assumed that $D_- = 0$ (i.e., the Stokes wave is resonant) and $D_+ \simeq 0$. Neglecting D_+ in Eq. (9) requires $|2k_+ a_+^{-1} \partial_\tau a_+| \gg |D_+|$. Therefore, the RFS instability will be in the four-wave resonant regime provided $|a_0|^2 |\zeta|/\tau \gg 8(k_p/k_0)^4$. For sufficiently long times, this condition will no longer be satisfied, and the RFS instability will transition into the four-wave nonresonant regime.

3.2. Four-wave nonresonant regime

In the weakly-coupled ($|\partial_\zeta \chi| \ll |k\chi|$) four-wave nonresonant regime, Eqs. (8) - (10) can be combined to yield

$$\left(\frac{\partial^3}{\partial \zeta \partial \tau^2} + \Gamma_{\text{4loc}}^2 \frac{\partial}{\partial \tau} \right) \chi \simeq -i\Gamma_N^3 \left[1 + \Delta_e \left(\frac{1}{2} + \frac{\zeta}{L} \right) \right]^{-4} \chi, \quad (16)$$

where we have kept the lowest-order term assuming $|D_+ a_+ / (2k_+ \partial_\tau a_+)| < 1$. The relativistic temporal growth rate of the RFS in the four-wave nonresonant regime without

chirp is

$$\Gamma_N = \left(\frac{k_p^7 |a_0|^2}{16 \gamma_{\perp 0}^{11/2} k_0^4} \right)^{1/3}. \quad (17)$$

We will assume that $|\Gamma_4^2 \partial_\tau \chi| \ll |\Gamma_N^3 \chi|$ such that the RFS interaction has moved from the four-wave resonant to the four-wave nonresonant regime and we may neglect the second term on the left-hand side of Eq. (16). Assuming the additional initial condition $\partial_\tau \chi(\tau = 0, \zeta) = 0$, Eq. (16) can be solved asymptotically and has the solution

$$\chi(\tau, \zeta) \sim \chi_0 \exp \left[\frac{3}{4} (\sqrt{3} + i) \Gamma_{\text{Neff}} (2|\zeta| \tau^2)^{1/3} \right], \quad (18)$$

where the effective growth rate is

$$\Gamma_{\text{Neff}} = \Gamma_N \left(\frac{L}{3\Delta_e |\zeta|} \right)^{1/3} \left\{ - \left(1 + \frac{\Delta_e}{2} \right)^{-3} + \left[1 + \frac{\Delta_e}{2} \left(\frac{1}{2} + \frac{\zeta}{L} \right) \right]^{-3} \right\}^{1/3}. \quad (19)$$

For $|\Delta_e| \ll 1$, the lowest-order correction to the growth rate due to chirp is $\Gamma_{\text{Neff}} \simeq \Gamma_N [1 - (2\Delta_e/3)(1 - |\zeta|/L)]$.

In deriving Eq. (18) it was assumed that $|\Gamma_4^2 \partial_\tau \chi| \ll |\Gamma_N^3 \chi|$. Using Eq. (18), this condition reduces to $|\zeta|/\tau \ll 2(\gamma_{\perp 0}/a_0^2)(k_p/k_0)^2$. Therefore, for sufficiently long times, the RFS will transition from the four-wave resonant regime ($|\Gamma_4^2 \partial_\tau \chi| \gg |\Gamma_N^3 \chi|$) to the four-wave nonresonant regime ($|\Gamma_4^2 \partial_\tau \chi| \ll |\Gamma_N^3 \chi|$). For longer times, the RFS instability can transition, for $a_0 \ll 1$, into the three-wave regime where the anti-Stokes wave can be neglected, or, for $a_0 \gtrsim 1$, into the strongly-coupled four-wave nonresonant regime.

3.3. Three-wave regime

In the three wave regime, we assume that the anti-Stokes wave is sufficiently out of resonance such that $|D_+ a_+| \gg |2k_+ \partial_\tau a_+|$. In this regime the anti-Stokes term may be neglected, and Eqs. (8) and (10) can be combined to yield

$$\frac{\partial^2 \chi}{\partial \zeta \partial \tau} \simeq -\Gamma_3^2 \left[1 + \Delta_e \left(\frac{1}{2} + \frac{\zeta}{L} \right) \right]^{-1} \chi, \quad (20)$$

where

$$\Gamma_3 = \frac{k_p^{3/2} |a_0|}{4 \gamma_{\perp 0}^{7/4} k_0^{1/2}} \quad (21)$$

is the relativistic temporal growth rate of the RFS instability in the three-wave regime without chirp. The solution to Eq. (20) (for $-L \leq \zeta \leq 0$) is

$$\chi(\tau, \zeta) = \chi_0 H(\tau) I_0 \left[2\Gamma_{3\text{eff}} \sqrt{\tau |\zeta|} \right], \quad (22)$$

where the effective growth rate of RFS in the three-wave regime is

$$\Gamma_{3\text{eff}} = \Gamma_3 \left\{ \frac{L}{\zeta \Delta_e} \ln \left[1 + \Delta_e \frac{\zeta}{L} \left(1 + \frac{\Delta_e}{2} \right)^{-1} \right] \right\}^{1/2}. \quad (23)$$

For $|\Delta_e| \ll 1$, the lowest-order correction to the growth rate due to chirp is $\Gamma_{3\text{eff}} \simeq \Gamma_3 [1 - (\Delta_e/4)(1 - |\zeta|/L)]$.

In deriving Eq. (20), the anti-Stokes wave was neglected $|D_+ a_+| \gg |2k_+ \partial_\tau a_+|$. Using Eq. (22) this condition for the three-wave RFS regime yields $|\zeta|/\tau \ll 16/(a_0^2 \gamma_{\perp 0}^{1/2})(k_p/k_0)^5$. In addition, the weakly-coupled approximation $|\partial_\zeta \chi| \ll |k\chi|$, assumed in Secs. 3.1 - 3.3, will no longer be valid at long times for sufficiently intense laser pulses $a_0 \gtrsim 1$. Assuming the perturbation grows in the resonant four-wave regime, the weakly-coupled approximation implies $|\zeta|/\tau \gg (k_p/k_0)^2 a_0^2 / (\gamma_{\perp 0}^3)$. For intense laser pulses at long times the RFS will violate this condition and transition into the strongly-coupled regime.

3.4. Strongly-coupled four-wave nonresonant regime

In the strongly-coupled $|\partial_\zeta \chi| \gg |k\chi|$ four-wave nonresonant $|\Gamma_4^2 \partial_\tau \chi| \ll |\Gamma_N^3 \chi|$ regime, Eqs. (8) - (10) can be combined to yield

$$\frac{\partial^4 \chi}{\partial \zeta^2 \partial \tau^2} \simeq 2k\Gamma_N^3 \left[1 + \Delta_e \left(\frac{1}{2} + \frac{\zeta}{L} \right) \right]^{-4} \chi, \quad (24)$$

for an flat-top pump laser pulse with a linear frequency chirp. The eikonal approximation $|\partial_\tau \chi| \ll |k\chi|$ is still assumed since the RFS instability enters the strongly-coupled four-wave nonresonant regime in the asymptotic limit $k_p \tau \gg 1$. The solution to Eq. (24) in the asymptotic limit can be approximated as

$$\chi \sim \chi_0 \exp \left[2^{5/4} (k\Gamma_N^3)^{1/4} \tau^{1/2} \left(\frac{L}{\Delta_e} \right)^{1/2} \left\{ \left[1 + \Delta_e \left(\frac{1}{2} + \frac{\zeta}{L} \right) \right]^{-1} - \left(1 + \frac{\Delta_e}{2} \right)^{-1} \right\}^{1/2} \right]. \quad (25)$$

For small chirp $|\Delta_e| \ll 1$, the RFS instability in the strongly-coupled four-wave nonresonant regime has the exponentiation $|\chi| \sim |\chi_0| \exp(N_{4\text{sc}})$, with

$$N_{4\text{sc}} \simeq 2^{5/4} (k\Gamma_N^3)^{1/4} (|\zeta|\tau)^{1/2} \left[1 - \frac{\Delta_e}{2} \left(1 - \frac{|\zeta|}{L} \right) \right]. \quad (26)$$

The RFS instability will be in the strongly-coupled four-wave nonresonant regime provided $16(k_p/k_0)^5 / (a_0^2 \gamma_{\perp 0}^{1/2}) \ll |\zeta|/\tau \ll (k_p/k_0)^2 a_0^2 / (8\gamma_{\perp 0}^3)$. This condition will be violated for sufficiently large pump laser intensities and propagation times, and the RFS instability will transition into the strongly-coupled three-wave regime.

3.5. Strongly-coupled three-wave regime

In the asymptotic limit $k_p \tau \gg 1$, the RFS instability will enter the strongly-coupled three-wave regime. In the asymptotic strongly-coupled three-wave regime, $|\partial_\zeta \chi| \gg |k\chi|$, $|\partial_\tau \chi| \ll |k\chi|$, and the anti-Stokes wave is no longer in resonance; therefore Eqs. (8) - (10) can be combined to yield

$$\frac{\partial^3 \chi}{\partial \zeta^2 \partial \tau} \simeq -2ik\Gamma_3^2 \left[1 + \Delta_e \left(\frac{1}{2} + \frac{\zeta}{L} \right) \right]^{-1} \chi, \quad (27)$$

for an flat-top pump laser pulse with a linear frequency chirp. The asymptotic solution to Eq. (27) can be approximated as

$$\chi \sim \chi_0 \exp \left[\frac{3}{2^{2/3}} (\sqrt{3} - i) (k\Gamma_3^2 \tau)^{1/3} \left(\frac{L}{\Delta_e} \right)^{2/3} \times \left\{ \left(1 + \frac{\Delta_e}{2} \right)^{1/2} - \left[1 + \Delta_e \left(\frac{1}{2} + \frac{\zeta}{L} \right) \right]^{1/2} \right\}^{2/3} \right]. \quad (28)$$

For small chirp $|\Delta_e| \ll 1$, the RFS instability in the strongly-coupled three-wave regime has the exponentiation $|\chi| \sim |\chi_0| \exp(N_{3sc})$, with

$$N_{3sc} \simeq \frac{3^{3/2}}{2^{2/3}} (k\Gamma_3^2)^{1/3} (|\zeta|^2 \tau)^{1/3} \left[1 - \frac{\Delta_e}{6} \left(1 - \frac{|\zeta|}{L} \right) \right]. \quad (29)$$

3.6. Summary of RFS instability regimes

Table 1 summarizes the exponentiation [number of e-folds, $\chi \propto \exp(N)$] of the RFS instability for a laser pulse with linear frequency chirp ($\partial_\zeta^2 \phi_0 = \Delta_e/L$), for $|\Delta_e| \ll 1$, in the regimes: four-wave resonant (N_4), four-wave nonresonant (N_{4nr}), three-wave (N_3), strongly-coupled four-wave nonresonant (N_{4sc}), and strongly-coupled three-wave (N_{3sc}). In each regime the exponentiation is increased for positive chirp (red wavelengths at the head of the pulse, blue wavelengths at the tail, such that $\Delta_e < 0$).

Figure 1 shows schematically the regimes of the FRS instability in $(a_0, |\zeta|/\tau)$ parameter space for $k_0/k_p = 10$. Initially the instability is dominated by the four-wave resonant mode (region 1 of Fig. 1). As the propagation time increases, the instability transitions into the four-wave nonresonant regime (region 2 of Fig. 1). Note that the instability always enters the four-wave nonresonant regime before moving into the strongly-coupled regime. At later propagation times, the instability will transition into the three-wave regime (region 3 of Fig. 1) for nonrelativistic interactions $a_0 \ll 1$, or the strongly-coupled four-wave nonresonant regime (region 4 of Fig. 1) for relativistic interactions [$a_0^4/\gamma_{\perp 0}^{5/2} > 2^7 (k_p/k_0)^3$]. For sufficiently long times, provided the interaction has not become nonlinear, the RFS instability will transition into the strongly-coupled three-wave regime (region 5 of Fig. 1). We also note that, in the limit of no chirp $\Delta_e = 0$ and

TABLE 1. Raman forward scattering growth rates [number of e -folds, $\chi \propto \exp(N)$] in the four-wave resonant (N_4), four-wave nonresonant (N_{4nr}), three-wave (N_3), strongly-coupled four-wave nonresonant (N_{4sc}), and strongly-coupled three-wave (N_{3sc}) regimes, including frequency chirp for $|\Delta_e| \ll 1$.

Exponentiation:	Regime:
$N_4 \simeq \frac{1}{2^{1/2}} \frac{a_0}{\gamma_{\perp 0}^2} \left(\frac{k_p}{k_0}\right) k_p (\zeta \tau)^{1/2} \left[1 - \frac{\Delta_e}{2} \left(1 - \frac{ \zeta }{L}\right)\right]$	$2 \frac{\gamma_{\perp 0}}{a_0^2} \left(\frac{k_p}{k_0}\right)^2 \ll \frac{ \zeta }{\tau}$
$N_{4nr} \simeq \frac{3^{3/2}}{8} \frac{a_0^{2/3}}{\gamma_{\perp 0}^{11/6}} \left(\frac{k_p}{k_0}\right)^{4/3} k_p (\zeta \tau^2)^{1/3} \left[1 - \frac{2\Delta_e}{3} \left(1 - \frac{ \zeta }{L}\right)\right]$	$\frac{16}{a_0^2 \gamma_{\perp 0}^{1/2}} \frac{k_p^5}{k_0^5} \ll \frac{ \zeta }{\tau} \ll 2 \frac{\gamma_{\perp 0}}{a_0^2} \frac{k_p^2}{k_0^2}$ and $\frac{ \zeta }{\tau} \gg \frac{a_0^2}{8\gamma_{\perp 0}^3} \frac{k_p^2}{k_0^2}$
$N_3 \simeq \frac{1}{2} \frac{a_0}{\gamma_{\perp 0}^{7/4}} \left(\frac{k_p}{k_0}\right)^{1/2} k_p (\zeta \tau)^{1/2} \left[1 - \frac{\Delta_e}{4} \left(1 - \frac{ \zeta }{L}\right)\right]$	$\frac{a_0^2}{8\gamma_{\perp 0}^3} \frac{k_p^2}{k_0^2} \ll \frac{ \zeta }{\tau} \ll \frac{16}{a_0^2 \gamma_{\perp 0}^{1/2}} \frac{k_p^5}{k_0^5}$
$N_{4sc} \simeq \frac{1}{2^{1/4}} \frac{a_0^{1/2}}{\gamma_{\perp 0}^{3/2}} \left(\frac{k_p}{k_0}\right) k_p (\zeta \tau)^{1/2} \left[1 - \frac{\Delta_e}{2} \left(1 - \frac{ \zeta }{L}\right)\right]$	$\frac{16}{a_0^2 \gamma_{\perp 0}^{1/2}} \frac{k_p^5}{k_0^5} \ll \frac{ \zeta }{\tau} \ll \frac{a_0^2}{8\gamma_{\perp 0}^3} \frac{k_p^2}{k_0^2}$
$N_{3sc} \simeq \frac{3^{3/2}}{4} \frac{a_0^{2/3}}{\gamma_{\perp 0}^{4/3}} \left(\frac{k_p}{k_0}\right)^{1/3} k_p (\zeta^2 \tau)^{1/3} \left[1 - \frac{\Delta_e}{6} \left(1 - \frac{ \zeta }{L}\right)\right]$	$\frac{ \zeta }{\tau} \ll \frac{16}{a_0^2 \gamma_{\perp 0}^{1/2}} \left(\frac{k_p}{k_0}\right)^5$ and $\frac{ \zeta }{\tau} \ll \frac{a_0^2}{8\gamma_{\perp 0}^3} \left(\frac{k_p}{k_0}\right)^2$

for weakly relativistic laser intensities $a_0 < 1$, the four-wave resonant, four-wave nonresonant, three-wave, and strongly-coupled three-wave regime growth rates reduce to essentially those described previously [19, 20]. However, the strongly-coupled four-wave nonresonant regime is a new regime of RFS, which has not been previously analyzed. This new regime is only important for relativistic laser intensities.

In this work a 1D model was assumed. The 1D approximation will be valid if the transverse Laplacian $\nabla_{\perp}^2$ in the wave equation operator can be neglected compared to the evolution operator for the scattered daughter waves $2k_{\pm} \partial_{\tau}$ [cf, the left-hand side of Eq. (9)]. If we assume that the transverse gradient scales as the laser spot size $\nabla_{\perp} \sim 1/r_s$, then the 1D limit $|\nabla_{\perp}^2 a_{\pm}| \ll |2k_{\pm} \partial_{\tau} a_{\pm}|$ is valid provided $1/(k_p r_s)^4 \ll [a_0^2/(2\gamma_{\perp 0}^4)] |\zeta|/\tau$ in the four-wave resonant regime. This condition can be rewritten as $1 \ll (4/\gamma_{\perp 0}^2) [2(P/P_c)(k_p/k_0)(k_p|\zeta|)(Z_R/\tau)]^{1/2}$, where $Z_R = k_0 r_s^2/2$ is the laser Rayleigh length and $P_c = k_p^2 a_0^2 r_s^2/32$ is the critical power for relativistic self-guiding. As may be expected, for typical laser-plasma parameters (e.g., $P/P_c \sim 1$, $k_0/k_p \sim 10$, and $k_p L \sim 10 - 100$), the 1D model will no longer be valid for propagation distances longer than a Rayleigh length $\tau > Z_R$.

FIGURE 1. Regimes of RFS instability [in parameter space $(a_0, |\zeta|/\tau)$ for $k_0/k_p = 10$]: (1) four-wave resonant, (2) four-wave nonresonant, (3) three-wave, (4) strongly-coupled four-wave nonresonant, and (5) strongly-coupled three-wave.

3.7. RFS chirp asymmetry

As Table 1 indicates, the spatiotemporal growth rate in all regimes of RFS is larger for a positively-chirped ($\Delta_e < 0$) than for a negatively-chirped ($\Delta_e > 0$) laser pulse. We may examine the asymmetry between laser pulses with positive and negative chirp by considering the ratio of the energy in the RFS generated plasma wave for the chirped to unchirped pump laser pulse

$$\frac{\mathcal{E}(\tau; \Delta_e)}{\mathcal{E}(\tau; 0)} = \frac{\int_L |\chi(\Delta_e)|^2 d\zeta}{\int_L |\chi(0)|^2 d\zeta}. \quad (30)$$

Figure 2(a) shows the plasma wave energy excited in the four-wave resonant regime by a laser pulse with a linear frequency chirp normalized to the unchirped excitation energy Eq. (30) versus propagation time $k_p \tau$, for $a_0 = 1$, $k_0/k_p = 10$ and $k_p L = 40$. Figure 2(b) shows the ratio of the plasma wave energy excited by positive and negative chirped pulses $\mathcal{E}(-|\Delta_e|)/\mathcal{E}(|\Delta_e|)$ versus chirp $|\Delta_e|$ after propagating $k_p \tau = 100, 200$, and 300 , for $a_0 = 1$, $k_0/k_p = 10$ and $k_p L = 40$. Figure 2 illustrates the relatively small influence of the frequency chirp on plasma wave generation. These results are also consistent with recent experimental observations, as discussed in Sec. 4.

4. DISCUSSION AND SUMMARY

In this paper, we have presented a calculation of the spatiotemporal growth of the Raman forward scattering instability produced by a frequency-chirped laser pulse propagating in an underdense plasma using the relativistic Maxwell-fluid equations. It was shown

FIGURE 2. (a) The plasma wave energy excited by a frequency chirped laser pulse normalized to the unchirped excitation energy versus propagation time $k_p\tau$ for several chirps $\Delta_e = -0.1, -0.05, 0, 0.05$, and 0.1 , with $a_0 = 1$, $k_0/k_p = 10$ and $k_pL = 40$. (b) The ratio of the plasma wave energy excited by positive and negative chirped pulses $\mathcal{E}(-|\Delta_e|)/\mathcal{E}(|\Delta_e|)$ versus chirp $|\Delta_e|$ after $k_p\tau = 100, 200$, and 300 , with $a_0 = 1$, $k_0/k_p = 10$, and $k_pL = 40$.

that a frequency chirp correlated to the longitudinal position within the laser pulse alters the exponentiation of the RFS instability. Table 1 summarizes the RFS growth rates in various regimes of the RFS instability. In particular, it was shown that positive chirped pulses (red wavelengths at the head of the pulse and blue wavelengths at the tail of the pulse) have a larger growth rate than negative chirped pulses (blue wavelengths at the head and red wavelengths at the tail).

The RFS instability is initially seeded by plasma density fluctuations or pump laser intensity fluctuations which contain Fourier components at the relativistic plasma frequency $k = k_p/\gamma_{\perp 0}^{1/2}$. For example the seeding of the RFS may be generated by thermal fluctuations in the plasma, ionization-induced plasma waves (owing to a time-varying

dielectric) [22, 23, 24], or ponderomotively-excited plasma waves (owing to the laser intensity gradient) [25, 26]. Experiments often will use the same pump laser pulse which undergoes self-modulation to create the plasma through ionization of a gas. Typically photoionization will occur very early in the head of the laser pulse, where the laser electric field becomes sufficiently intense such that the rate of ionization is maximum [27]. This will create a plasma density front moving with the laser. The amplitude of the ionization-induced plasma waves created by this ionization front will be approximately $\chi_0 \sim a_0^2(\zeta_{\text{ionz}})/4$, where ζ_{ionz} is the location in the pump laser pulse where the laser electric field is sufficiently intense to ionize the background gas. Note that the ionization location ζ_{ionz} is weakly dependent on the chirp. For typical laser experimental parameters, the shift in the ionization location is less than a laser wavelength $\zeta_{\text{ionz}} < \lambda_0$. The amplitude of the ponderomotively-excited plasma waves scales as $\chi_0 \sim a_0^2/(k_p L)^2$, where the gradient in the laser pulse intensity is $\sim 1/L$. For short laser pulses, the ponderomotively excited plasma wave will typically dominate other sources for seeding the RFS instability.

Previous theoretical work by Dodd and Umstadter [15] on Raman scattering of a chirped laser pulse used a heuristic calculation of the group velocity dispersion to estimate the effect of a linear frequency chirp. Dodd and Umstadter claimed that, in the nonrelativistic regime ($a_0 \ll 1$), the amount of chirp necessary to completely eliminate the Raman scattering instability is $\Delta\omega_0 = (\omega_0/\omega_p)\gamma_R/2$ where $\gamma_R = (\omega_p/\sqrt{8})(\omega_p/\omega_0)a_0$ is the usual unchirped temporal growth rate. This predicts a complete elimination of plasma wave generation through RFS by using a bandwidth of $\Delta\omega_0/\omega_0 \simeq 1.8\%$ for the parameters $a_0 = 1$ and $\omega_0/\omega_p = 10$. Furthermore PIC simulations of Dodd and Umstadter imply stabilization of RFS using a 20% bandwidth for the parameters $a_0 = 1$ and $\omega_0/\omega_p = 10$. In contrast, the analytic solutions to the Maxwell-fluid equations presented in Sec. 3 show only a modest change in the RFS growth rate. We find, for a 20% negative chirp ($\Delta_e = 0.2$), the growth rate in the four-wave resonant regime at the center of the pulse is reduced by only 4.7% compared to the unchirped growth rate.

Recent experiments [12, 13] using long laser pulses ($k_p L \gg 1$) with a frequency chirp $|\Delta_e| \approx 3\%$ operating in the self-modulated laser wakefield regime have measured RFS growth independent of the chirp. These experiments are consistent with the calculations presented in Sec. 3 where a 3% chirp produces little asymmetry [cf, Sec. 3.7]. Experiments operating in the regime $k_p L \sim 1$, i.e., the standard laser wakefield accelerator regime, have reported asymmetry in the measured Stokes wave [13] and electron yield [14]. We believe this asymmetry in the standard laser wakefield regime is due to preferential seeding of the RFS instability by asymmetric pulse envelopes generated by nonlinear contributions to the chirp in the laser compression system [14].

In conclusion, the spatiotemporal growth of the plasma wave generated by RFS was calculated using the relativistic Maxwell-fluid equations for a laser pulse with a linear frequency chirp in various regimes. It was shown that the growth rate of RFS increases (decreases) for positive (negative) chirp. In addition, we have shown that a linear frequency chirp with $|\Delta_e| \ll 1$ produces only a small effect on the growth of the RFS instabilities, and therefore will have only a minor effect on the enhancement or suppression of Raman instabilities for laser-plasma applications.

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