Parametric Scattering Instabilities in Inhomogeneous Plasmas*

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Raman and Brillouin scatterings in inhomogeneous and blowoff plasmas are analyzed. For normal incidence, the threshold for near-90° Raman scattering is a minimum, necessitating the analysis without the WKB approximation. Pump depletion may severely limit the laser penetration into a plasma.

Recently, there has been considerable interest in the possibility of inducing nuclear fusion by laser irradiation of a deuterium pellet. Essential to its feasibility is the question of the nature and efficiency of the laser-plasma coupling. Among the basic processes of the laser-plasma interaction is the parametric excitation of the collective modes in the plasma by the intense laser light, leading to possible anomalous absorption[1] as well as anomalous scattering.[2] As the back-scattering by the plasmon or the ion wave (Raman and Brillouin scattering) has maximum growth rate, the underdense region of the plasma becomes reflective rather than transparent to the incident light, once a threshold power is reached. In an inhomogeneous plasma, the matching condition for the resonant three-wave interaction holds only in a limited region in the direction of the inhomogeneity and the propagation of the decay waves out of this "resonant region" of interaction then limits the growth, leading to possible stabilization of the parametric process. With the usual WKB approximation, the maximum amplification has been shown to be $\exp(2\pi\gamma_0^2/V_1V_2K')$, where γ_0 is the growth rate in a homogeneous plasma, V_1 and V_2 are the group velocities of the decay waves along the gradient in x, and $K'=(d/dx)[k_{0x}(x)-k_{1x}(x)-k_{2x}(x)]$ is the rate of change of the mismatch.[3] This greatly raises the threshold power needed for Raman back-scattering for incidence normal to the density gradient because of the fast propagation of the back-scattered light out of the interaction region.[4] Recent computer simulation[5] with a one-dimensional model has verified this threshold power and demonstrated the importance of the back-scattering process. However, for the scattered light propagating almost perpendicular to the density gradient, its convection out of the resonant region along the gradient is slow and a lower threshold power for the side-scattering is thus expected. For the side-scattering, the usual WKB approximation for the light wave is not valid, necessitating the present analysis. Here, the principal limitation on the growth is due to the refraction of the side-scattered wave out of the resonant region. For normal incidence, the threshold power for Raman side-scattering is $18T_e/mc^2$ lower than that of the back-scattering and is comparable to that of the Brillouin back-scattering. Consider a light wave $\vec{\epsilon}_0=2\epsilon_0\vec{e}_0\sin(k_0x-\omega_0t)$ normally incident on a plasma with a density profile $n(x)=n_0(1+x/L)$. The coupled equations for the Raman scattering are

$$3v_e^2\nabla^2n_e-\frac{\partial^2n_e}{\partial t^2}-\omega_p^2\left(1+\frac{x}{L}\right)n_e=-2\left(\frac{e}{mc}\right)^2\frac{\epsilon_0}{k_0}\cos\varphi\,\nabla^2[\cos(k_0x-\omega_0t)A], \tag{1}$$

$$c^2\nabla^2A-\frac{\partial^2A}{\partial t^2}-\omega_p^2\left(1+\frac{x}{L}\right)n_e=2\,\frac{\omega_p^2c\epsilon_0}{\omega_0n_0}\,n_e\cos\varphi\cos(k_0x-\omega_0t), \tag{2}$$

where n_e is the electron density fluctuation and $\vec{A}=\vec{e}_2A$ is the vector potential for the scattered light; $\omega_p^2=4\pi n_0e^2/m$, $\cos\varphi=\vec{e}_0\cdot\vec{e}_2$, $v_e=(2T_e/m)^{1/2}$ is the electron thermal speed. For side-scattering, the plasma wave absorbs the incident momentum. Letting

$$n(x,t)=\tilde{n}(x,t)\exp\{i[\textstyle\int^x k_{1x}(x)\,dx+\vec{k}_{1\perp}\cdot\vec{r}_\perp-\omega_1t]\},\quad A(x,t)=\tilde{A}(x,t)\exp\{-i[\vec{k}_{1\perp}\cdot\vec{r}_\perp+\omega_2t]\},$$

where

$$\omega_1=[\omega_p^2(x)+3k^2(x)v_e^2]^{1/2},\quad \omega_2=[\omega_p^2(0)+k_\perp^2c^2]^{1/2}=\omega_0-\omega_1,\quad k_{1x}(0)=k_0(0),$$

we have, from Eqs. (1) and (2)

$$\frac{\partial \tilde{n}}{\partial t} + V_g \frac{\partial \tilde{n}}{\partial x} = i \frac{n_0 \epsilon_0}{2\omega_p} \left(\frac{e}{mc}\right)^2 \frac{k^2}{k_0^2} \cos\varphi \, \tilde{A} \exp(iK'x^2/2), \tag{3}$$

$$-\frac{2i\omega_2}{c^2} \frac{\partial \tilde{A}}{\partial t} + \frac{\partial^2 \tilde{A}}{\partial x^2} - \frac{\omega_p^2}{c^2 L} x\tilde{A} = \frac{\omega_p^2 \epsilon_0}{\omega_0 n_0 c} \tilde{n} \cos\varphi \exp(-iK'x^2/2), \tag{4}$$

where

$$K' = (d/dx)[k_0(x) - k_{1x}(x)] = (\omega_p^2/6Lv_e^2)[k_{1x}^{-1} + (ev_e^2/c^2)k_0^{-1}].$$

Because the scattered light wave is near its turning point, there is a rapid phase shift due to refraction; hence we neglect the plasma-wave phase shift, validly if

$$K' x_{\text{int}}^2 \approx \tfrac{1}{3}(\omega_p/\omega_0)^{2/3}(v_e/v_0)^{4/3}(k_0 L)^{-1}(c/v_e)^2 \ll 1, \tag{5}$$

where $x_{\text{int}} = (Lc^2/\omega_p^2\gamma)^{1/3}$ is the width of the interaction region where the wave growth takes place; $\gamma = \frac{1}{8}(v_0/v_e)^2 k_0 L \cos^2\varphi \ll 1$ is the exponential amplification factor. Equation (5) is approximately satisfied for $k_0 L \gtrsim 10^3$, with typical laser-fusion parameters. The present calculation neglects the stabilizing effect of the finite radius R of the laser beam[6] and is justified for $R \gg k_0 x_{\text{int}}^2$, or $R/L > (\omega_0/\omega_p)^{4/3} \times \gamma^{-2/3}(k_0 L)^{-1/3}$. Combining Eqs. (3) and (4) and adding a source term for the steady state, we have

$$d^3A/dx^3 - \alpha\, d(xA)/dx - i\alpha\gamma A = \delta(x), \tag{6}$$

where $\alpha = \omega_p^2/c^2 L$. The boundary conditions, corresponding to the physical situation of the scattering process, are such that at $x \to \infty$, there are a spatially damped electromagnetic wave and an outgoing plasma wave; at $x \to -\infty$ there is only the outgoing (scattered) electromagnetic wave. The solution to the homogeneous equation can then be obtained by Fourier transformation of Eq. (6) and has the following integral representation:

$$\begin{aligned} A_+(x) &= g\int_{C_1} dk\, f(k) + h\int_{C_3} dk\, f(k), \quad \text{for } x > 0, \\ A_-(x) &= \int_{C_2} dk\, f(k) - \int_{C_1} dk\, f(k), \quad \text{for } x < 0, \end{aligned} \tag{7}$$

where $f(k) = \exp[i(-\frac{1}{3}k^3 - kx + \gamma \ln k)]$ and the contours (C_1, C_2, C_3) are from 0 to ∞ along $[\exp(-i\pi/6), \exp(i\pi/2), \exp(i7\pi/6)]$; g and h are determined by the requirements that $A_+(x)$ and $A_-(x)$ as well as their first derivatives match at $x = 0$. At large x, the behavior is given by the contributions from the saddle points at $-i(\alpha x)^{1/2}$ and γ/x for $A_+(x)$ and at $(\alpha|x|)^{1/2}$ for $A_-(x)$ with the following form:

$$A_+(x) = \tfrac{1}{2} g\sqrt{\pi}(\gamma/x)^{i\gamma - 1/2} + h\sqrt{\pi}(\alpha/x)^{1/4}\exp(-\tfrac{2}{3}\sqrt{\alpha}x^{3/2}), \quad x \to \infty,$$

$$A_-(x) = \sqrt{\pi}(\alpha/|x|)^{1/4}\exp(-i\tfrac{2}{3}\sqrt{\alpha}|x|^{3/2}), \quad x \to -\infty.$$

We note the first term in A_+ together with $\exp(ik_0 x)$ constitutes an outgoing electron plasma wave; the other terms are the usual Airy function for the electromagnetic wave near its turning point. Thus, the above solution satisfies the proper boundary conditions. At $x = 0$, A_+ and A_- can be evaluated in terms of Γ functions:

$$\begin{aligned} A_+(0) &= \tfrac{1}{3}(3\alpha)^{(1+i\gamma)/3}\Gamma((1+i\gamma)/3)\{g\exp[\tfrac{1}{6}\pi(\gamma - i)] + h\exp[(-7\pi/6)(\gamma - i)]\}, \\ A_-(0) &= \tfrac{1}{3}(3\alpha)^{(1+i\gamma)/3}\Gamma((1+i\gamma)/3)\{\exp[-\tfrac{1}{2}\pi(\gamma - i)] - \exp[\tfrac{1}{6}\pi(\gamma - i)]\}. \end{aligned} \tag{8}$$

Similarly, $A_+'(0)$ and $A_-'(0)$ can be obtained. Setting $A_+(0) = A_-(0)$ and $A_+'(0) = A_-'(0)$, we find $g = -\{1 + \exp[-(2\pi\gamma/3)]\}$, $h = -\exp(2\pi\gamma/3)$. The solution to the inhomogeneous equation, Eq. (6), is

$$A(x) = [A_+(x)\theta(x) + A_-(x)\theta(-x)]/[A_-''(0) - A_+''(0)],$$

where $\theta(x)$ is the step function. The jump in A'' at $x = 0$ can readily be found to be

$$A_+''(0) - A_-''(0) = -i(3\alpha)^{(1+i\gamma)/3}\Gamma(1 + i\gamma/3)\exp(-\pi\gamma/2).$$

The amplitude at $x = 0$ with a unit source is therefore

$$A(0) = -\frac{\exp(2\pi\gamma)^{13}}{3(3\pi)^{2/3}}\frac{\Gamma((1+i\gamma)/3)}{\Gamma((3+i\gamma)/3)}[\exp(\tfrac{1}{2}\pi i) + \exp(-2\pi\gamma/3)]. \tag{9}$$

Amplification over the thermal source is therefore $A^2 = I\exp[(2\pi/9)(v_0/v_e)^2 k_0 L \cos^2\varphi]$. The threshold for Raman side-scattering is therefore $\frac{1}{9}(v_0/v_e)^2 k_0 L > 1$, much lower than that of backscattering: $(v_0/c)^2 k_0 L > 1$.

For Brillouin scattering by ion waves, Eq. (3) is replaced by

$$\left(\frac{\partial}{\partial t} + \frac{k_x}{k} C_s \frac{\partial}{\partial x}\right) n_e = i \frac{n_0 m k}{2 C_s M k_0} \left(\frac{e}{mc}\right)^2 \cos(\varphi) \tilde{A} \exp(\tfrac{1}{2} i K' x^2), \tag{10}$$

where $C_s = (T_e/M)^{1/2}$ is the sound speed. The phase shift of the ion wave is principally due to the nonuniform expansion velocity $U(x)$ in a blowoff plasma,[7] in addition to the temperature and density gradients, with the result

$$K' = \frac{d}{dx}(k_0 - k_{1x} - k_{2x}) = 2k_0 L_u^{-1} \sin^2\left(\frac{\theta}{2}\right)\left\{1 + \frac{2L_u}{L_t}\sin^2\left(\frac{\theta}{2}\right) + \left(\frac{\omega_p}{\omega_0}\right)^2\left(\frac{L_u}{2L_n}\right)\left[(\cos\theta)^{-1} + 4\left(\frac{\omega_0}{\omega_p}\right)^4\left(\frac{v_e}{c}\right)^2\right]\right\},$$

where $L_u^{-1} = -d\ln(U)/dx$, $L_T^{-1} = d\ln(T_e)/dx$, and $L_n^{-1} = d\ln(n)/dx$, and the gradient of U is assumed to be opposite to that of density and temperature. Typically, in a blown-off plasma, $L_n < L_u \ll L_T$ because of rapid electron heat conduction and hydrodynamics of the expanding plasma. A temporally growing (absolute) instability exists for the scattering angles θ_i at which $K' = 0$.[4] These angles are $\theta_1 \approx 0$ (forward scattering) and $\theta_2 \approx \cos^{-1}[-(\omega_p^2/2\omega_0^2)L_u/L_n]$ for negligible v_e^2/c^2 and L_n/L_T. The growth rate of these absolute instabilities, $\gamma_{\rm eff} \approx \omega_{pi}(v_0/c)|\cos\theta_i|^{-1/2}$, is much less than the maximum growth rate of the convective modes, which is the same as the growth rate in a homogeneous plasma: $\gamma_0 = (\omega_{pi}/2)(v_0/C_s) \times (k_1 C_s/k_0 c)^{1/2}$. For experiments of short duration τ_d, the absolute instability is important only for $\gamma_{\rm eff}\tau_d \gg 1$. Otherwise convective modes are dominant. We therefore examine the amplification of the convective modes with $K' \neq 0$. With the usual WKB approximation, we find

$$A^2 = I\exp\left\{\frac{\pi}{4}\left(\frac{v_0}{v_e}\right)^2\left(\frac{\omega_p}{\omega_0}\right)^2 k_0 L_u \cos^2\varphi\left[\sin^2\left(\frac{\theta}{2}\right)\left\{\cos\theta + \frac{\omega_p^2}{2\omega_0^2}\frac{L_u}{L_n}\left[1 + 4\cos\theta\left(\frac{\omega_0}{\omega_p}\right)^4\left(\frac{v_e}{c}\right)^2\right]\right\} + \frac{L_u}{2L_T}\cos\theta\right]^{-1}\right\}. \tag{11}$$

A^2 tends to peak both for $\cos\theta \approx (L_u/2L_n)(\omega_p/\omega_0)^2$ and $\theta \approx 0$. For nearly forward scattering, the WKB approximation for the ion wave breaks down for $\sin^2(\theta/2) \lesssim \frac{1}{4}k_0 L_T)^{1/2}$. The analysis without the WKB approximation can similarly be carried out yielding the amplification over the thermal source: $E_k^2 = I_k \exp[(8\pi/3)(v_0/v_e)^2(\omega_p/\omega_0)^2 k_0 L_T \cos^2\varphi]$. The forward scattering essentially converts the coherent incident light into incoherent, scattered light in the same direction.

As a result of the stimulated scattering, the pump (incident wave) energy is depleted at a rate given by the energy-conservation equation:

$$(c/8\pi)\partial E_0^2/\partial x + \int d^3k \langle 2\Gamma_k I_k \rangle = 0, \tag{12}$$

where $\Gamma_k = [\gamma_0^2 - \frac{1}{4}(\omega_0 - \omega_1 - \omega_2)^2]^{1/2}$ is the growth rate in a homogeneous plasma with finite frequency mismatch, which is also the maximum growth rate of the decay wave in an inhomogeneous plasma. $2\Gamma_k I_k = \tau^{-1} KT \int_0^\tau dt\, 2\Gamma_k \exp(2\Gamma_k t) \approx 2KT\Gamma_k G^{-1}$; $\exp(G)$ is the mean rate of increment of the decay wave energy until the maximum gain G is reached either by inhomogeneity or by nonlinearity. For Raman side-scattering in inhomogeneous plasmas,

$$G = (2\pi/9)\Gamma_k^2 (c/v_e)^2 L \cos^2(\varphi)/\omega_p c \xrightarrow[\Delta\omega\to 0]{} (2\pi/9)(v_0/v_e)^2 k_0 L \cos^2\varphi,$$

as given by Eq. (9). Integrating Eq. (12), we obtain the variation of the pump intensity due to the Raman scattering dominated by side-scattering:

$$\exp[-(2\pi/9)\eta(x)k_0 L] - \exp[-(2\pi/9)\eta(0)k_0 L] \approx (2\pi^2 k_0^3 n_c^{-1})\eta^{-1/2}(0)(v_e/c)^{1/2}(\omega_p/\omega_0)^{5/3}(k_0 L)^{-1/3} x/L, \tag{13}$$

where $\eta(x) = E_0^2(x)/8\pi n T_e$, n_c is the critical density at which $\omega_p = \omega_0$, and the Landau damping becomes negligible for $x > 0$, i.e., $k_0\lambda_D(0) \lesssim 0.2$. The penetration depth of the laser light, l, defined by $\eta(l) = \frac{1}{2}\eta(0)$ is then found to be

$$\frac{l}{L} = \frac{n_c}{2\pi^2 k_0^3}\left(\frac{c}{v_e}\right)^{1/2}\left(\frac{\omega_0}{\omega_p}\right)^{5/3}(k_0 L)^{1/3}\eta^{1/2}(0)\exp\left[-\frac{\pi}{9}\eta(0)k_0 L\right]. \tag{14}$$

For parameters typical of a plasma corona in the laser-pellet irradiation experiment, $k_0 = 2\pi \times 10^4$

(Nd-glass laser), $k_0L \simeq 10^3-10^5$, and $(c/v_e)^2 \approx 10^3$, l/L is less than unity for $k_0L\eta(0) = 70$, provided that other nonlinear effects have not set in to further limit the growth.

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Temperature-Induced Explosive First-Order Electronic Phase Transition in Gd-Doped SmS

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We have found a reversible temperature-induced explosive first-order electronic phase transition, with decreasing temperature at atmospheric pressure, in $Sm_{1-x}Gd_xS$ for $0.16 \leq x < 0.22$. The shiny golden-yellow metallic crystal disintegrates to a black powder. The transition is isostructural, but the unit cell expands from $a \approx 5.68$ to 5.82 Å. The Sm ion in the black phase is characterized by an intermediate valence of ~2.45. We present a phase diagram showing the stability regions in the T-x plane and discuss a plausible driving mechanism for the transition.

A pressure-induced first-order isostructural semiconductor-to-metal transition due to $4f$-electron delocalization was recently reported in pure SmS at 6.5-kbar pressure at room temperature.[1] With a view to lowering this transition pressure we studied the effect of Gd substitution for the Sm ion in SmS. Since the lattice constant of GdS is substantially smaller than that of SmS, 5.563 Å compared to 5.97 Å for pure SmS, Gd substitution may be expected to have the same effect as applying pressure. Accordingly, we find that Gd substitution lowers the transition pressure and, further, that concentrations in excess of 15 at.% Gd stabilize the "collapsed" metallic phase at atmospheric pressure. The lattice parameter abruptly decreases to 5.68 Å near this concentration, and the samples exhibit a golden-yellow metallic reflectivity. We have discovered that this metallic phase when cooled exhibits an explosive first-order phase transition at ambient pressure in which the bright shiny sample disintegrates into a black powder. The black phase has the NaCl-type structure and is characterized by an intermediate valence of 2.45 for the Sm ion due to a partial localization of the electrons on the $4f$ band of Sm. We believe that the driving force for the transition is the following: The $4f$ level of Sm moves towards the Fermi level when temperature is lowered, resulting in fractional occupation of the level. This causes expansion of the lattice, which in turn increases the overlap of the $4f$ level with the Fermi level. This bootstrapping drives the transition.

Samples were made by reacting appropriate amounts of Sm and Gd with S in an evacuated quartz tube at 500°C for 15 h, followed by heat treatment at 900°C for 2 h, and then melting the reacted material in a sealed evacuated tantalum tube. The material thus prepared was well crystallized and appeared black in color when the Gd concentration was ≤ 15 at.%. At higher concentrations of Gd the samples have a metallic golden-yellow luster. X-ray powder photographs were taken to obtain the lattice parameters and to con-