Ionization Induced Scattering of Short Intense Laser Pulses

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Intense laser pulses propagating in gases undergoing ionization are subject to a scattering instability due to the dependence of the ionization rate on the laser electric field. The instability is convective, and growth is limited for a pulse of finite extent by propagation out of the unstable region. In the nonlinear regime, where the scattered wave amplitude becomes large, the scattering instability saturates at a level that gives rise to full modulation of both the plasma density and laser pulse amplitude.

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Recent progress in the development of ultraintense, short pulse lasers has stimulated interest in the study of the interaction of intense electric fields with gases and plasmas. At power densities greater than $10^{13}$ to $10^{14}$ W/cm$^2$, which are easily achieved and exceeded with today’s lasers, pulses propagating in gas rapidly ionize atoms creating plasmas which strongly modify the index of refraction. With the terawatt lasers now being used in a wide range of plasma experiments this intensity threshold can be achieved well before the beam reaches its focus. This ionization process leads to a number of interesting nonlinear phenomena, including frequency upshifting of the laser radiation by the moving ionization front [1–7], refractive defocusing of the laser pulse due to the radial inhomogeneity of the plasma electron density [8,9], and harmonic generation due to the nonlinear dependence of the ionization rate on the field amplitude [8].

An additional effect that has received only a small amount of attention is the possible scattering of the radiation by the collective amplification of modulations of the electron density transverse to the initial direction of propagation of the laser pulse [10–12]. Such a transversely modulated density appears in the presence of transverse modulations of the laser amplitude due to the dependence of the ionization rate on field amplitude. The modulations of electron density scatter the laser wave, which can reinforce the modulations in field amplitude and lead to instability. The purpose of this Letter is to examine this phenomenon for the conditions expected for short pulse lasers. We will investigate the instabilities in both the linear and the nonlinear regimes, and determine the effect of finite pulse extent and duration on their development.

We begin by considering the simple case of laser propagation in a background gas of atomic hydrogen. (Results from more refined models will be presented as well.) The situation is then described by the wave equation for the laser electric field $\mathbf{E}$ along with the rate equation for the production of plasma electrons,

$$\left(\frac{1}{c^2} \frac{\partial^2}{\partial t^2} + \nabla \times \nabla \times \right) \mathbf{E} = -k_p^2 \frac{n}{n_g} \mathbf{E} \quad (1)$$

$$\frac{\partial}{\partial t} \frac{n}{n_g} = \nu(|\mathbf{E}|) \left(1 - \frac{n}{n_g}\right) \quad (2)$$

Here $n$ is the plasma electron density, $n_g$ is the uniform constant background gas density, $\nu(|\mathbf{E}|)$ is the field dependent ionization rate, for example, due to tunneling ionization [13], and $k_p^2 = 4\pi e^2 n_g/mc^2$ is the plasma wave number in the case where the gas is totally ionized.

Analysis of these equations proceeds by examining the linear stability of a plane wave pulse to perturbations with transverse modulations. Specifically, we write the laser electric field in terms of a normalized amplitude $a = e\mathbf{E}/(mc^2k_0)$. We then express this amplitude and electron density as the sum of a plane wave “equilibrium” $a_0(z, \xi) = ct - z$, $n_0(z, \xi)$ with the characteristic wave number $k_0$, and perturbations $\delta a(z, \xi)$ and $\delta n(z, \xi)$ with the characteristic wave number $k_\perp$ in the transverse direction $x$, $a = a_0 \hat{a} \exp(-ik_0 \xi) + \text{c.c.}$, where

$$\hat{a} = [a_0 + \delta a_+ \exp(ik_\perp x) + \delta a_- \exp(-ik_\perp x)], \quad (3a)$$

$$n = n_0 + \delta n \exp(ik_\perp x) + \delta n^* \exp(-ik_\perp x). \quad (3b)$$

Note that our equilibrium actually depends on time and space in a nontrivial way due to the ionization of the gas as the finite duration laser pulse propagates through it. The shape of the pulse is determined by its dependence on the laser frame coordinate $\xi = ct - z$. In the limit of a tenuous plasma, the pulse evolves slowly as it propagates in $z$. We insert expressions (3a) and (3b) into (1) and (2), linearized with respect to the perturbations, and average over one period of the laser field. (This last step requires that the ionization rate be much less than the laser frequency. This will later be found to be the case of interest for the scattering instability.) We then obtain coupled equations describing the equilibrium and the perturbation,

$$2 \frac{\partial}{\partial z} \left( -ik_0 + \frac{\partial}{\partial \xi} \right) a_0(\xi, z) = -k_p^2 \frac{n_0}{n_g} a_0(\xi, z), \quad (4a)$$

$$c \frac{\partial}{\partial \xi} \frac{n_0(\xi, z)}{n_g} = \nu(|a_0|) \left(1 - \frac{n_0(\xi, z)}{n_g}\right). \quad (4b)$$

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$2 \frac{\partial}{\partial z} \left( \frac{\partial}{\partial \xi} + ik_0 \right) \delta a_{\pm} + \left( k_p^2 \frac{n_0}{n_g} + k_\xi^2 \right) \delta a_{\pm} = -k_p^2 \frac{\delta n}{n_g} e^{(\nu)} \right), \quad (5a)$

c $\frac{\partial}{\partial \xi} \frac{\delta n}{n_g} = -\nu(|a_0|) \frac{\delta n}{n_g} + c k_1 a_0 \delta a_{\pm} + a_0 \delta a |a_0|^2. \quad (5b)$

Here, $\nu(|a_0|) =\int k_0 d\xi \nu(|a_0| \exp(-ik_\xi) + c.c.)/2\pi$ is the cycle averaged ionization rate for the pump pulse, and

$$k_1 = \frac{|a_0|}{2c} \frac{\partial \nu}{\partial |a_0|} \left[ 1 - \frac{n_0}{n_g} \right], \quad (6)$$

describes the coupling between the field and density perturbations.

Solutions to Eqs. (4a) and (4b), describing the evolution of a plane wave pulse, have been studied extensively. A sample solution illustrating their character is shown in Fig. 1, where the laser field amplitude and electron density are shown for the case of a 100 fs, full width at half maximum, $1.4 \times 10^6$ W/cm$^2$ maximum power pulse propagating in 100 Torr of helium. The laser wavelength is taken to be 0.8 $\mu$m. In obtaining these solutions, Eq. (4b) has been generalized in a straightforward way to treat the multiple ionization stages of helium. Figure 1(a) shows that the plasma which envelopes the body of the laser pulse slows down its propagation, causing it to fall back in the laser frame coordinate $\xi = ct - z$. Figure 1(b) shows that the helium is ionized in two stages. Early in the pulse's propagation the ionization occurs rapidly, resulting in two sharp steps in the electron density. As the pulse propagates the ionization occurs more gradually due to the fact that the portions of the pulse in different densities of plasma propagate with different group velocities and separate. In particular, the head of the pulse propagates at the speed of light while the body of the pulse, which is in plasma, has a group velocity $v_g = c(1 - k_p^2/k_0^2)^{1/2}$.

Turning our attention now to the perturbations, we note that the coupling between the radiation and density perturbations is nonzero when the ionization rate depends on field amplitude and the gas (or a particular ionization state) is only partially ionized. Thus, the instability, if present, will exist during the short times when the pump field is large enough to ionize but has yet to completely ionize a particular charge state, that is, during the “riser” portion of the density staircase in Fig. 1(b). As a first step in analyzing this process we neglect momentarily the time and space dependence of the equilibrium and solve Eqs. (5a) and (5b) by assuming the coefficients are constants. The result is a dispersion relation [10] for perturbations with an assumed time and space dependence of the form $\exp[i(k_\xi z - k_\xi \xi)]$.

$$0 = D(k_\xi, k_\xi) = \frac{\nu}{c} - ik_\xi + k_1 \left[ \frac{k_p^2}{k_\xi^2 + 2k_\xi(k_\xi + k_0)} + \frac{k_p^2}{k_\xi^2 + 2k_\xi(k_\xi - k_0)} \right]. \quad (7)$$

If one regards the temporal-like wave number $k_\xi$ as fixed (and much smaller than the laser wave number $k_0$) and solves for the spatial-like wave number $k_\xi$, one obtains

$$k_\xi = \pm \frac{1}{2k_0} \sqrt{k_\xi^4 + 2k_\xi^2 \frac{k_1 k_p^2}{\nu/c - ik_\xi}}. \quad (8)$$

Note that, if $k_\xi \ll \nu/c$, Eq. (8) has the same form with regard to the perpendicular wave number dependence as that of a self-focusing instability. In this case, instability is possible only if the coupling wave number $k_1$ is negative; that is, the ionization rate decreases with field amplitude. This instability would correspond to self-focusing in which the index of refraction decreases where
the laser field is small. Typically, however, \( k_1 \) is positive, and instability occurs only for perturbations with nonzero temporal variation in the pulse (i.e., \( k\xi \neq 0 \). In the limit \( k\xi = \bar{\nu}/c = k_1 \), and \( k_{\perp} \gg k_p \), we obtain, for the spatial growth rate,

\[
\text{Im}[k_z] = \pm \frac{k_1k_z^2k_\xi}{2k_0[k_1^2 + (\bar{\nu}/c)^2]}.
\]  

(9)

This has a maximum spatial growth rate which, for \( k_1 = \bar{\nu}/c \), is approximately \( \text{Im}[k_z] = k_2^p/k_0 \). For the usual self-focusing instability due to a nonlinear dielectric constant \( n_2I \), Eq. (8) is modified with \( k_1k_z^2/(\bar{\nu}/c - ik_\xi) \) replaced by \(-k_0^2n_2I\). In this case, the maximum growth rate is \( \text{Im}[k_z] = k_0n_2I/2 \), and occurs for \( k_{\perp}^2 = k_0^2n_2I \). (On the other hand, the growth for the ionization scattering instability increases monotonically with \( k_\perp \).

The ionization scattering instability will dominate when \( k_\perp \) is due to the propagation of the scattered electromagnetic frame, perturbations are only convectively unstable. This will occur when \( k_1 \) replaces by \(-k_0^2n_2I\). In this case, the maximum growth rate is \( \text{Im}[k_z] = k_0n_2I/2 \), and occurs for \( k_{\perp}^2 = k_0^2n_2I \). (On the other hand, the growth for the ionization scattering instability increases monotonically with \( k_\perp \).

The use of formulas such as (8) and (9) is limited since the temporal-like wave number \( k_\xi \) must be determined self-consistently. To this end, we determine the impulse response [14] to a disturbance initiated at \( \xi = z = 0 \) by simultaneously solving \( D(k_z + k_\xi z, k_\xi) = 0 \) and \( \partial D(k_z + k_\xi z, k_\xi)/\partial k_\xi = 0 \). The amount of complex exponentiation \( \Gamma = i(k_z z - k_\xi) \) is then found in the limit \( k_{\perp} \gg k_p \) in which a single pole is kept in Eq. (6) to be

\[
\Gamma = -\frac{\bar{\nu}}{c} \left( \xi - \frac{k_1^2}{2k_0^2} z \right)
\]

\[
\pm (1 + i)k_p\sqrt{\frac{k_1}{k_0}} z \left( \xi - \frac{k_2^p}{2k_0} z \right),
\]

(10)

where it is assumed \( \xi > zk_1^2/(2k_0^2) \) (otherwise there is no response). From (10) it is clear that, at a fixed point \( \xi \) in the pulse, the disturbance will grow and then decay as the pulse propagates and \( z \) increases. That is, in the pulse frame, perturbations are only convectively unstable. This is due to the propagation of the scattered electromagnetic wave which has a group velocity in the \( z \) direction \( v_g = c[1 - k_1^2/2k_0^2] \) and therefore falls behind the initial laser pulse. If one maximizes (10) over \( \xi \) for fixed \( z \), one finds \( \text{Re}[\Gamma_{\text{max}}(z)] = zk_2^p/k_1/[4k_0(\bar{\nu}/c)] \) which is equivalent to (9) with \( k_\xi = \bar{\nu}/c \).

The above estimates are only qualitative since the time and space dependence of the equilibrium cannot be ignored. In particular, the coupling term \( k_1 \) which is responsible for the instability will be nonzero during only a portion of the laser pulse of thickness \( \delta z = c/\bar{\nu} \) while the gas is ionizing. This is comparable to the wavelength of the most unstable perturbation. Additionally, the region of ionization evolves as the pulse propagates due to the lower-than-light speed of the pulse in plasma. For these reasons, quantitative predictions on the growth of perturbations require the solution of differential equations in the form of (5). A solution of these equations (again for the case of helium) is shown in Fig. 1(b). In these calculations the scattered waves were taken to have an initial spatial dependence of the form \( \delta a_{zz} = \delta a_0 \sin(\pi \xi/L) \), where \( L \) is the length of the pulse corresponding to a duration of 100 fs and \( \delta a_0 \) in this case, similar to that of the equilibrium depicted in Fig. 1. Plotted is the amount of exponentiation \( [\delta a_{zz}/\delta a_0] \) for the scattered waves for the case \( k_{\perp}/k_p = 0.8 \). As can be seen, as a function of \( \xi \), growth is positive only in those regions where the gas is undergoing ionization from one stage to another. As the pulse propagates, these regions grow in size and the amount of growth increases, reaching a level of \( 10^3 \) after only 0.6 cm of propagation.

The growth depicted in Fig. 1(b) is for a specific choice of transverse wave number. Figure 2 shows the maximum amount of growth (maximized over \( \xi \)) as a function of transverse wave number for several propagation distances. The wave number which gives maximum growth corresponds to \( k_{\perp} = k_p \). This is not a resonance phenomenon, but rather, it follows from a competition between the local growth and convection of the scattered waves out of the unstable region. According to (8) the local growth rate must vanish as \( k_{\perp} \to 0 \). For \( k_{\perp} \gg k_p \) the local growth rate becomes independent of \( k_{\perp} \) according to (9). Thus the local growth is a monotonically increasing function of \( k_{\perp} \).

Because of the convective nature of the growth, Eq. (10) indicates that the disturbances propagate out of the unstable region of \( \xi \) more rapidly with increasing \( k_{\perp} \).

With a value of \( k_{\perp} = k_p \) the disturbances propagate backward through the pulse at about the same rate that the pulse is stretched due to the plasma’s reduction of the equilibrium group velocity.

To examine the nonlinear regime of the scattering instability, we solve the system (1) and (2) generalized to

\[
\text{FIG. 2. Scattered wave growth maximized over } \xi \text{ as a function of the scattered wave number for several propagation distances.}
\]

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multiple ionization stages for parameters corresponding to those of Figs. 1 and 2. The wave equation is treated in the envelope approximation assuming cylindrical symmetry, basically Eq. (4a) with the addition of the transverse Laplacian [15]. Furthermore, the susceptibility on the right-hand side of (4a) includes the linear excitation of the plasma wave, the relativistic self-focusing nonlinearity, and the nonlinear susceptibility of the background gas. However, these latter effects are weak for the parameters under investigation. Figure 3 shows the results of a simulation corresponding to a 760 mJ, 100 fs FWHM pulse with a Gaussian radial profile of intensity focused to 200 µm in 100 Torr jet of helium. Plotted is the electron density immediately after the pulse has passed as a function of radius and axial distance. Because of refraction by the plasma, the pulse spreads. As it spreads it becomes unstable to the scattering instability. This is evidenced by the striations in the density shown in Fig. 3 (note, the radial and axial scales are different). The transverse wave number of the density modulations corresponds to the plasma wave number based on the local electron density. The laser pulse also does not remain smooth but develops transverse structure with 100% variations. Thus, the instability severely distorts the shape of the laser pulse and leaves, in the wake of the pulse, a nonuniform plasma consisting of long striations with transverse dimension on the order of the collisionless skin depth.

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