Demonstration of Collisionally Enhanced Degenerate Four-Wave Mixing in a Plasma

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Experimental evidence of degenerate four-wave mixing in a plasma is presented for the first time. The thermal-force contribution to the signal reflectivity, as evidenced by its temporal shape and dependence on plasma density, pump and probe intensities, and polarizations, dominates that due to the ponderomotive force in the collisional plasma. The pump-pump grating, which is driven by a much stronger ponderomotive force, is found not to affect adversely the signal reflectivity.

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It is well established that phase conjugation by degenerate four-wave mixing (DFWM) has applications in the areas of pointing and tracking, coherent image amplification, real-time adaptive optics, optical computing, and communications.[1] In DFWM, two counterpropagating pump and one probe beam of identical frequencies interact within the nonlinear optical mixing medium, to generate a backward (to the probe) traveling signal wave that is the phase conjugate of the probe beam. Although DFWM has been demonstrated in solids, liquids, and gases with use of both resonant as well as nonresonant media, no conclusive demonstration of this phenomenon in plasmas has been reported.

The phase-matching conditions for DFWM are $\omega_f+\omega_b-\omega_p-\omega_s=0$ and $\mathbf{k}_f+\mathbf{k}_b-\mathbf{k}_p-\mathbf{k}_s=0$, where f, b, p, and s denote forward, backward, probe, and signal (phase-conjugate) waves, respectively. In plasmas there are two dominant mechanisms that can generate the plasma-density gratings necessary for DWFM. These are the ponderomotive force resulting from the beating of each of the two pump waves with the probe wave[2] and the thermal force resulting from localized collisional heating.[3] It has recently been predicted that in plasmas were the collisional mean free path is shorter than the grating wavelength, the contribution to the signal reflectivity due to the thermal force can be greater than that due to the ponderomotive force. This thermal-force enhancement of the plasma-density perturbations can lead to thermal self-focusing which is of concern to laser fusion.[4]

The third-order susceptibility, χ_3, responsible for DFWM in a plasma is given by Federici and Mansfield[3] to be

$$\chi_3 \cong \frac{n_0/n_c^2}{8(4\pi)^2 T_e+T_i}\left[1+\sum_{i=f,b}\frac{n_0\nu_e}{(\mathbf{k}_i-\mathbf{k}_p)^2\kappa_e}\right]. \qquad (1)$$

The first term is the contribution to χ_3 due to the ponderomotive force whereas the second term is the additional contribution to χ_3 due to collisional heating. Here ν_e is the electron-ion collision frequency, κ_e is the electron thermal conductivity, $\mathbf{k}_i-\mathbf{k}_p$ is the wave vector of the plasma-density grating due to the interaction of the forward pump and the probe beams or the backward pump and the probe beams, n_0/n_c is the ratio of the plasma density to the critical density, and T_e and T_i are the electron and ion temperatures. The third-order polarization for the phase-conjugate signal wave resulting from the two gratings is given by

$$P_{3,s}=\chi_{3,fp}(E_fE_p^*)E_b+\chi_{3,bp}(E_bE_p^*)E_f. \qquad (2)$$

This nonlinear polarization results in a power reflectivity for the signal wave which, in the limit of small R, is given by[2]

$$R=I_s/I_p\cong\kappa^2L^2, \qquad (3)$$

where κ is the coupling coefficient given by $\kappa\cong|(2\pi\omega/c)\chi_3E_fE_b|$, and L is the interaction length. Note that R is independent of the probe intensity but is linearly related to the pump intensities.

An advantage of phase conjugation in a plasma by DFWM rather than by nearly DFWM at the nearby ion acoustic resonance[5] is that only in the former case are the gratings insensitive to density and temperature gradients, since they are stationary density perturbations and there is no ion Landau damping. In the limit of low signal reflectivities, pump depletion and oscillation effects can be neglected and the phase-conjugation process can be explained physically as follows: Each of the forward and backward pumps beat with the probe to set up two stationary ion-density perturbations in the plasma. The backward and the forward pumps Bragg scatter from these density perturbations, giving rise to the signal wave which is the phase conjugate of the probe wave. Indeed, substitution of the expression for χ_3 into the expression for the signal reflectivity shows that, in the limit of small R, we recover the familiar Bragg scattering formula for the grating reflectivity,[6]

$$R_{(f,b)p}=C\left[\frac{\pi}{2}\frac{n_1}{n_0}\frac{n_0}{n_c}\frac{L}{\lambda}\right]^2, \qquad (4)$$

where C is the ratio $I_{f,b}/I_p$ and n_1/n_0 is the density perturbation level of the grating given by

$$\left(\frac{n_1}{n_0}\right)_{\text{tot}} = \left(\frac{n_1}{n_0}\right)_{\text{ponderomotive}} (1+A)$$

$$= \sum_{i=f,b} \left(\frac{v_0}{c}\right)_i \left(\frac{v_0}{c}\right)_p \frac{1}{4} \frac{mc^2}{T_e+T_i}(1+A). \quad (5)$$

Here $v_0/c = eE/m\omega c$ is the normalized oscillating velocity of the electrons in the electromagnetic field and $A \cong n_0 \nu_e/(\mathbf{k}_i - \mathbf{k}_p)^2 \kappa_e$ is the thermal enhancement factor.

Substitution into Eq. (1) shows that even in a very underdense, collisionless ($n_0 < 10^{-2} n_c$), 1-eV plasma, $10^{-13} \leq \chi_3 \leq 10^{-4}$ esu for $10^{-3} \leq \lambda \leq 10$ cm. Thus, particularly at long wavelengths, the third-order plasma susceptibilities can be much larger than those for other currently available materials. Furthermore, the response time of the plasma grating can be much faster than that of other materials, such as the artificial Kerr media now being developed for DFWM in the gigahertz frequency range.[7] At lower frequencies still (5–50 MHz), the ionosphere provides a natural plasma source capable of retroreflecting the pump radiation, enabling phase conjugation via DFWM.

The experimental setup is shown in Fig. 1. A hybrid Co_2 TEA oscillator operating on the $10\text{-}P_{20}$ transition produces up to 300 mJ of energy in either a single longitudinal mode or a multimode, 150 ns (FWHM) long, gain-switched, TEM_{00} pulse. A 10% reflectivity beam splitter is used to generate the probe beam (p), whereas a 50% beam splitter is used to generate the two pump (backward b, and forward f) beams. A single 5-m radius-of-curvature focusing mirror (M) is used to bring all three beams to a common focus at the center of the plasma chamber. The focal spot size is about 1 mm. The beam intensities are varied by inserting attenuators

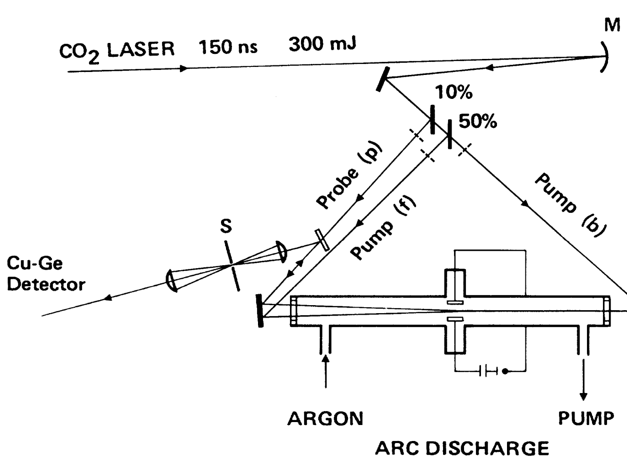

FIG. 1. Experimental setup for observing DFWM in a plasma.

after the beam splitters as shown by the dashed lines in Fig. 1. The two pump beams are counterpropagating and the probe beam propagates at an angle of 2.4° with respect to the forward pump beam. The probe beam overlaps the two pumps near the center of the plasma chamber over a length of about 1.5 cm. The grating wave numbers are $k_{fp} = 248$ cm^{-1} and $k_{bp} = 1.18 \times 10^4$ cm^{-1}.

The plasma is produced by a high-current arc discharge in 4 Torr of Ar gas by using 2.5-cm-long rail-gap electrodes. The plasma density is determined by measuring the Stark broadening of the 6563-Å hydrogen Balmer-α line by adding 10% H_2 to the Ar gas. The phase-conjugate signal is detected by using a liquid-helium–cooled Cu-Ge detector. A spatial filter (S) is used to reduce the stray-light level. For calibration purposes the plasma chamber can be removed and replaced with a hot CO_2 cell containing 100 Torr of CO_2 gas at 450 °C.[8] Phase conjugation is readily observed in this resonantly absorbing gas and enables us to check that our beams are aligned correctly. The maximum intensities of the pumps (10^8 W/cm^2) and the probe beam (10^7 W/cm^7) are too low for stimulated Brillouin scatter to occur. The stimulated-Brillouin-scatter threshold is experimentally found to be $\geq 10^{10}$ W/cm^2 for our conditions.[9] Furthermore, since we expect that the single-mode bandwidth of our laser is less than the ion acoustic frequency of the large- and small-k gratings, roughly $f_{bp} \sim 580$ MHz and $f_{fp} \sim 12$ MHz, we neglect any contribution to the signal wave due to nearly DFWM at the ion acoustic resonance.[5]

That the experimentally observed signal wave is indeed due to DFWM is confirmed in the following manner: First, when the forward and backward pumps and the probe wave have the same polarizations, the signal wave completely disappears if the probe or either of the two pump beams is blocked. Second, if the polarization of the probe beam is rotated by 90°, the signal wave again disappears because the orthogonally polarized beams can no longer form gratings. Also, the signal-wave ray retraces[10] the incident probe pulse. When half of the probe beam is masked, the signal wave appears only on the unmasked side of the probe, even though the pump beams that do the Bragg scattering are not masked.

The plasma density at the arrival of the laser pulse can be tuned over a limited range by varying the time of arrival of the laser pulse with respect to the arc. The measured signal reflectivity versus laser timing is shown in Fig. 2. The relative importance of the two gratings is readily established by rotating the polarizations of the pump beams. Recall from Eq. (5) that the ponderomotive contribution to n_1/n_0 is the same for both the large- and the small-k gratings. For $T_e = T_i = 4$ eV,[11] $(v_0/c)_{f,b} \cong 10^{-4}$, and $(v_0/c)_p \cong 3 \times 10^{-5}$, we obtain $(n_1/n_0)_{\text{ponderomotive}} \cong 4.1 \times 10^{-5}$. Collisional heating, however, is expected to make a significant contribution

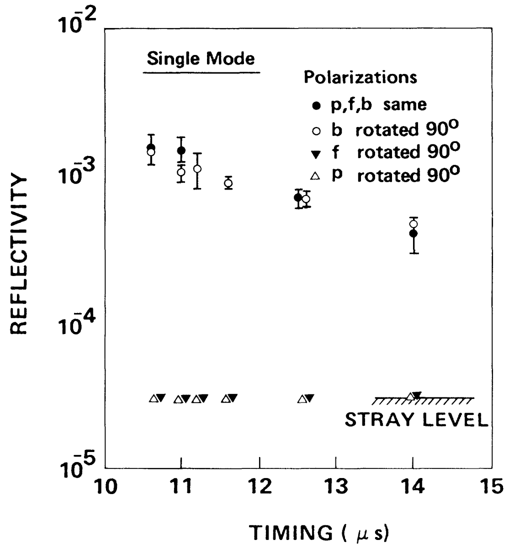

FIG. 2. Variation of signal reflectivity vs laser timing with respect to the arc for various combinations of polarizations of the f, b, and p beams. $I_{f,b}=100$ MW/cm^2, $I_p=10$ MW/cm^2.

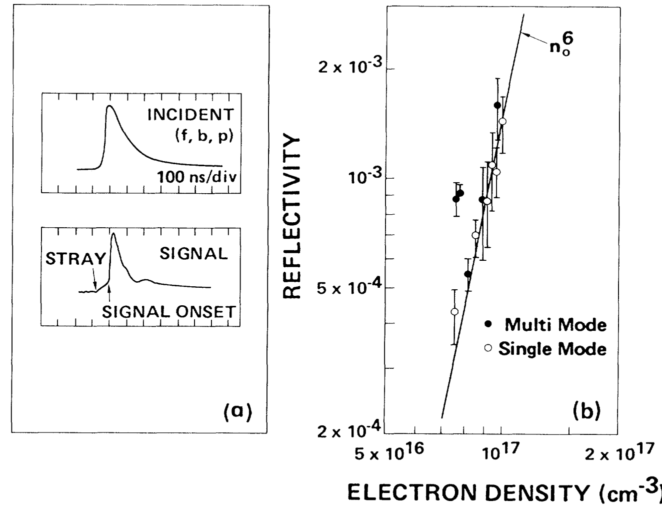

FIG. 3. (a) Temporal shapes of the incident (f, b, and p) and signal (s) beams, and (b) signal reflectivity vs plasma density. $I_{f,b}=100$ MW/cm^2, $I_p=10$ MW/cm^2.

only to the small-k grating since the electron mean free path, λ_{ei}, of 10 μm (for $n_0=10^{17}$ cm^{-3}, $T_e=4$ eV) is much smaller than the grating wavelength of 253 μm. As can be seen from Fig. 2, when the polarization of the forward pump is rotated by 90°, the signal reflectivity drops dramatically to below the measurement threshold. However, when the polarization of the backward pump is rotated by 90°, the signal reflectivity is almost the same as when all three beams (f, b, and p) have the same polarization. Furthermore, the polarization of the signal beam is found to be the same as that of the backward pump beam under all circumstances. It is therefore clear that most of the signal reflectivity is due to the small-k grating formed by the interference of the forward pump and the probe beams. The level of density perturbation can be estimated from the measured signal reflectivity to be $n_1/n_0 \sim 4.7\times10^{-4}$ at an arc timing of 11 μs. Thus, the thermal enhancement factor, A_{expt}, is approximately 10, which is in reasonable agreement with the value calculated directly, $A_{\text{theory}} \sim 6.3$, by substituting for ν_e and κ_e in Eq. (5). This accounts for the observed reflectivity being dominated by the small-k grating. Using Eqs. (1), (4), and (5) we calculate χ_3 to be 1.5×10^{-12} esu for a measured plasma density of 9.5×10^{16} cm^{-3} at 11-μs laser timing and $T_e=T_i=4$ eV.

Figure 2 also shows that the pump-pump grating, which is driven by a much stronger ponderomotive force than the pump-probe gratings, does not adversely affect the signal reflectivity.[12] When the two pumps are orthogonally polarized (b rotated by 90°, Fig. 2), the pump-pump grating cannot form and yet the signal reflectivity is the same as when all three beams have the same polarization.

That the major contribution to the reflected signal was from the small-k grating was further confirmed by comparing the temporal shapes of the signal and the pump beams. The characteristic growth and decay time of each grating term which contributes to the nonlinear polarization [Eq. (2)] should be on the order of a few times $(c_s k_{fp,bp})^{-1}$, where c_s is the ion acoustic speed. For the small-k grating this is >13 ns, whereas for the large-k grating the characteristic growth time is >0.3 ns. Figure 3(a) shows the experimentally observed signal roughly 50 ns after the onset of the pump, which further substantiates that the signal reflectivity is due to the small-k grating.

With use of Eqs. (3) and (5) it is easy to show that if the signal reflectivity is dominated by the thermal force it should scale as

$$R \propto \lambda^{10} n_0^6 I_f I_b L^2 / T_e^{10}. \tag{6}$$

We plotted the measured signal level versus plasma density for both single-mode and multimode laser operation [Fig. 3(b)]. The reflectivity due to the small-k grating, in the measured density range, indeed scales as $\sim n_0^6$. Recall that the signal reflectivity due to the ponderomotive force alone scales as n_0^2. This further confirms that the thermal force is enhancing the small-k grating. The variation of the signal reflectivity with pump and probe intensities is shown in Fig. 4. As expected, the signal reflectivity was found to be nearly independent of the probe intensity ($R \propto I_p^{0.14}$). When the pump power is increased, however, the signal reflectivity begins to saturate after the initial linear increase with intensity, as ex-

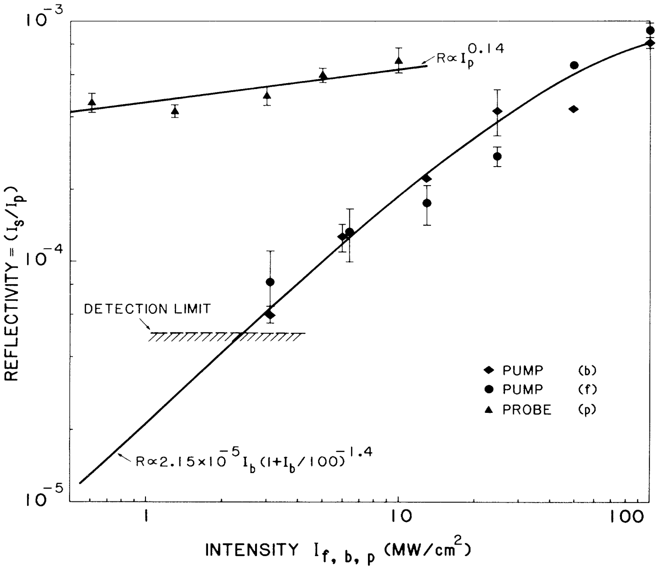

FIG. 4. Signal reflectivity (R) vs pump and probe intensities.

pected. Since most of the signal reflectivity is due to the thermal force, whose dependence on temperature is $1/T_e^{10}$, the increase in the electron temperature, when the intensity of one of the pumps is increased from 5 MW/cm^2 to $\sim$100 MW/cm^2 (a net increase in total laser power of a factor of 2), accounts for the departure from linear dependence on laser power. A reasonable fit to the observed variation of reflectivity with pump powers is obtained assuming the dependence

$$R = 2.15\times10^{-5} I_b (1 + I_b/I_f)^{-1.4}, \qquad (7)$$

which for $I_f = 100$ MW/cm^2 shows a linear growth at low intensity followed by a gradual saturation at higher intensity when temperature effects become important (Fig. 4).

In conclusion, we have observed DFWM in a plasma for the first time. The signal reflectivity is almost completely due to the small-k grating. This experiment has demonstrated the importance of collisional effects in allowing the thermal enhancement of the plasma-density grating. The pump-pump grating is found not to adversely affect the signal reflectivity. Since $\chi_3 \propto \lambda^{10}$, collisional plasmas ($\lambda_{\mathrm{mfp}} \ll \lambda_{\mathrm{grating}}$) appear to be extremely promising for phase conjugation via DFWM in the microwave regime.

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