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"Studies of Degenerate and Nearly Degenerate Four Wave

Mixing of Laser Radiation in Plasmas"

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1 Introduction

Optical Phase Conjugation is an area of nonlinear optics with a wide variety of potential applications. One method of generating a phase conjugate signal is with four wave mixing (FWM). In FWM, three input beams interact in a nonlinear medium, and a fourth beam is produced that is the phase conjugate of one of the input waves. Degenerate Four Wave Mixing (DFWM) is a special case of FWM in which all of the beams are at the same frequency. In a plasma, DFWM is an effective technique for phase conjugation in high density, low temperature plasmas. One way of enhancing the phase conjugate signal over and above the DFWM level is with Resonant Four Wave Mixing (RFWM), in which two of the input beams beat at a plasma resonance. In addition to enhancing the generated wave, RFWM can also serve as a diagnostic for many plasma parameters, such as the electron and ion temperatures, the ion acoustic velocity, and the damping rate. In this report, experimental evidence of RFWM with CO₂ laser radiation (10.6 μ m) is presented, and the data is compared with theoretical predictions.

2 Experimental Set-up

The experimental set-up is shown in Figure 1. The CO₂ laser produces a gain switched, single longitudinal mode, 150 nanosecond FWHM pulse, with

Figure 1: Experiment Set-Up

an energy of about about 200 millijoules. After leaving the oscillator, a telescope is used to expand the beam from a width of 0.7 cm to 3.5 cm. The main beam is then split into three, first with a 10% beam-splitter, and then with a 50% beam-splitter. The three beams are focused, and converge in a 1 mm spot in the center of the plasma. The two strong pump beams are counter-propagating, while the weaker probe beam enters at a slight angle with respect to the forward pump. The plasma is produced by a high current arc discharge in a few torr of gas (Argon, Nitrogen, Helium or Hydrogen). Some typical parameters for the plasma are a density $n_0 = 10^{17} \text{ cm}^{-3}$, and electron and ion temperatures $T_e = T_i = 4 \text{ eV}$. The phase conjugate signal is generated at the center of the plasma, and retraces the path of the probe beam. The signal is then split with a 50% beam-splitter, and its intensity is measured with a liquid-helium cooled, Cu:Ge detector. A retro reflecting mirror can be placed in the path of the probe beam to simulate the phase conjugate signal. This also provides a reference intensity for the probe beam. The reflectivity is defined as the ratio of signal intensity to the probe intensity.

3 Degenerate Four Wave Mixing

3.1 Ponderomotive Force

Four Wave Mixing is a nonlinear process that is dependent on the material's third order susceptibility, $\chi^{(3)}$. In a plasma, the susceptibility is related to the density modulations produced by the laser radiation. In the case of DFWM, there are two mechanisms that can produce these density modulations : the ponderomotive force and the thermal force. The ponderomotive force is proportional to the gradient of the laser's intensity, and produces a periodic modulation in the density. The third order susceptibility $\chi^{(3)}$ due to the ponderomotive force has been calculated by Steel and Lam :

$$\chi^{(3)} = \frac{n_0/n_c^2}{(8\pi)^2(T_e + T_i)} \quad (1)$$

Here, n_0 is the plasma electron density, $n_c = m\omega^2/4\pi e^2$ is the critical density of the plasma, and $T_e(T_i)$ is the plasma electron (ion) temperature. The theoretical value for the reflectivity is given by

$$R = \left| \frac{2\pi\omega}{c} \chi^{(3)} I L \right|^2, \quad (2)$$

where I is the laser intensity and L is the interaction length. If $\chi^{(3)}$ is determined by the ponderomotive force, then the reflectivity should scale as

$$R \propto \frac{n_0^2 \lambda^6}{T_e^2}. \quad (3)$$

Another way of visualizing four wave mixing in a plasma is by using a grating picture. Two of the waves interfere to form a density grating, and the third beam Bragg scatters off this grating. For example, in Figure 2, the forward pump and probe interfere to form a stationary, long wavelength grating, and the backward pump scatters off this. Similarly, the backward pump and probe form a stationary, short wavelength grating for the forward pump. The reflectivity can be rewritten with the Bragg Scattering formula :

$$R = C \left| \frac{\pi}{2} \frac{n_0}{n_c} \frac{n_1}{n_0} \frac{L}{\lambda} \right|^2, \quad (4)$$

where C is the ratio of the pump intensity to the probe intensity, and the density perturbation level is

$$\frac{n_1}{n_0} = \sum_{i=f,b} \left(\frac{v_o}{c} \right)_i \left(\frac{v_o}{c} \right)_p \frac{1}{8} \frac{mc^2}{T_e + T_i}, \quad (5)$$

where $v_o/c = eE/m\omega c$ is the normalized oscillating velocity of the electrons.

3.2 Thermal Force

The two gratings have equal importance when the ponderomotive force is the nonlinear mechanism. When the thermal force is significant, however, this is no longer the case. Since the plasma heats in proportion to the laser intensity, the laser produces a temperature grating in the plasma. This temperature modulation then produces a density grating to supplement the ponderomotive force. The effects of this thermal force are significant only when the collisional mean free path is shorter than the density grating scale length. Therefore, the thermal force is more important for the small k (long wavelength) grating. The density perturbation n_1/n_0 now becomes

$$\frac{n_1}{n_0} = \sum_{i=f,b} \left(\frac{v_o}{c} \right)_i \left(\frac{v_o}{c} \right)_p \frac{1}{8} \frac{mc^2}{T_e + T_i} (1 + A), \quad (6)$$

Small k ,
Large λ

FORMATION

Large k ,
Small λ

READOUT

$D = \lambda \sin \theta$

$D = \lambda / 2 \cos(\theta/2)$

Figure 2: Gratings

where $A = n_0 \nu_e / k_p^2 \kappa_e$ is the thermal enhancement factor. The reflectivity now scales as

$$R \propto \frac{n_0^6 \lambda^{10}}{T_e^{10}}. \quad (7)$$

Experimental evidence of DFWM was reported in Kitigawa et al. The experiments showed that the reflectivity scales as n_0^6 , which shows the importance of the thermal force. By rotating the polarization of the input beams, the importance of the small k grating was also demonstrated.

4 Resonant Four Wave Mixing

4.1 Theory

The density fluctuations of the plasma gratings, and therefore the reflectivity, of four wave mixing can be further enhanced by the method of resonant four wave mixing. In RFWM, the pump beams have identical frequencies w_0 , while the probe beam has a slightly different frequency $w_p = w_0 + w_m$, where $w_m \ll w_0$. The frequency difference between the pumps and the probe is chosen so that $w_0 - w_p = kc_s$, where k is the grating wavenumber, and c_s is the ion acoustic velocity. When this condition is met, the density modulation is increased by the generation of the ion acoustic wave, whose dispersion relationship is given by

$$w = k_{ion} c_s. \quad (8)$$

For temperatures of $T_e = T_i = 2.25$ eV, the ion acoustic velocity in an argon plasma is equal to

$$c_s = \sqrt{\frac{Z T_e + \gamma T_i}{M}} = 4.64 \times 10^5 \text{ cm/s}. \quad (9)$$

The wavenumber for the small k grating for a probe angle of 4° is $k_{fp} = 413.74 \text{ cm}^{-1}$, so the resonant frequency $w_{res} = 192$ MHz, or $f_{res} = w_{res} / 2\pi = 30.57$ MHz. If the frequency of the probe beam is shifted with respect to the forward pump by 30.57 MHz, the ion acoustic wave can be excited in the plasma.

In our experiment, the frequency shift is produced by acousto-optically modulating the single mode CO₂ laser pulse. A transducer sends a acoustic

wave through a germanium crystal operating in a standing wave mode. The laser scatters off this wave, and the light in the first diffracted order is both upshifted and downshifted by the acoustic frequency. This now becomes the probe beam. The light in the zeroth order is not greatly affected by the acoustic wave, and serves as the pump beam. Using this set-up, the frequency of the probe beam can be shifted from 20 MHz to 60 MHz.

An estimate of the increase of the density modulation n_1/n_0 can be made with a simple oscillator model. From the equations of motion and continuity, the equation for the ion acoustic wave in the presence of an external force, in steady state, is given by

$$(w^2 + jw\gamma - k^2 c_s^2)n_1 = \frac{jkF_{NL}}{M}. \quad (10)$$

In this equation, γ is the damping rate, and F_{NL} is assumed to be the ponderomotive force.

From the definition of the ponderomotive force,

$$F = \frac{-e^2}{2mw^2} \nabla \cdot F^2, \quad (11)$$

the right hand side of equation 10 can be rewritten as

$$\frac{-jk}{M} F_{NL} = \frac{mc^2 k^2 c_s^2}{2(T_e + T_i)} \left(\frac{v_o}{c}\right)_f \left(\frac{v_o}{c}\right)_p. \quad (12)$$

From Equation 5, this becomes

$$\frac{-jk}{M} F_{NL} = \left(\frac{n_1}{n_0}\right)_{pond} \cdot k^2 c_s^2. \quad (13)$$

Therefore, the total density modulation (in steady state) is equal to

$$\left(\frac{n_1}{n_0}\right)_{tot} = \left(\frac{n_1}{n_0}\right)_{pond} \times \frac{k^2 c_s^2}{w^2 + jw\gamma - k^2 c_s^2} \quad (14)$$

When the frequency difference is equal to the ion acoustic frequency, the real part of the denominator vanishes, and the grating strength is equal to

$$\left(\frac{n_1}{n_0}\right)_{tot} = \left(\frac{n_1}{n_0}\right)_{pond} \times \left(\frac{w}{j\gamma}\right). \quad (15)$$

If $\nu_{ii} \gg kc_s$, then collisional effects, which give rise to a thermal force, are important. When these thermal forces are included, the density modulation becomes

$$\left(\frac{n_1}{n_0}\right)_{tot} = \left(\frac{n_1}{n_0}\right)_{pond}(1 + A) \times \left(\frac{w}{j\gamma}\right) \quad (16)$$

Since the reflectivity from Equation 4 is proportional to $(n_1/n_0)^2$, the reflectivity in resonant four wave mixing should be increased by a factor of $(w/\gamma)^2$. In addition, since the right hand side of Equation 14 is Lorentzian in form, the reflectivity as a function of frequency difference should have a Lorentzian shape, where the width of the profile is related to the damping rate, γ .

4.2 Experimental Results

Resonant four wave mixing experiments were performed in three gases, Argon, Nitrogen, and Helium. The first order diffracted beam from the acousto-optic crystal was used for the probe beam, while the zeroth order was used for the pump beams. The pump beams have a strong component at w_0 and small components at $w_0 \pm w_m$, while the probe beam has strong components at $w_0 \pm w_m$ and a weak component at w_0 . Typical pulse shapes of the probe and signal beams are shown in Figure 3. For the probe beam, the beating of the frequencies at $w_0 + w_m$ and $w_0 - w_m$ produce an intensity modulation in the pulse at a frequency $w = 2w_m$.

For Argon, the probe beam enters the chamber at an angle of 4° with respect to the forward pump. The frequency applied to the crystal is varied from 20 MHz to 35 MHz. At each frequency, the peak reflectivity is measured over several shots, and the average peak reflectivity is plotted in Figure 4. The plot shows a significant peak at about 30.5 MHz, and then gradually decreases. Away from resonance, the reflectivity should return to the DFWM value. The enhancement due to RFWM seems to be approximately 4. The resonance peak is very narrow, with a FWHM of about 2 MHz.

The experiment was then performed with a Nitrogen plasma. The resonance frequency is proportional to the ion acoustic velocity, c_s , which has a $1/\sqrt{M}$ dependence. Since the mass of Nitrogen is smaller than the mass of Argon by a factor 14/40, the resonant frequency is expected to be greater by $\sqrt{40/14} = 1.69$. The plot of reflectivity vs. frequency for Nitrogen shows a peak at about 50 MHz, with a width of about 6 MHz. The ratio of the

25 0.57 1146

5.12

25 0.57 1146

Figure 3: Typical Pulse Shapes

Argon, Experimental Data

Figure 4: Experimental Data

resonant frequency of Nitrogen to the resonant frequency of Argon is

$$\frac{f_{res}(N_2)}{f_{res}(Ar)} = \frac{50\text{MHz}}{30.5\text{MHz}} = 1.64 \quad (17)$$

The shift in resonance frequency agrees very well with the theoretical prediction.

The experiment was also performed with a Helium plasma. Since Helium has a much smaller mass than Argon, the frequency shift should be very large. Due to the limits of the crystal modulation frequency, the angle between the probe and the forward pump was reduced from 4° to 2° . This reduces the wavenumber k , and therefore reduces the resonance frequency. The peak of the Helium reflectivity curve is at about 49 MHz. Comparing this with the theoretical prediction,

$$\frac{f_{res}(He)}{f_{res}(Ar)} = \frac{49\text{MHz} \times 2}{30.5\text{MHz}} = 3.21 \quad (18)$$

The mass ratio is $\sqrt{40/4} = 3.16$, so this also agrees very well with the theory. In Figure 5, the three resonant frequencies are plotted, along with a theoretical $1/\sqrt{M}$ dependence.

In order to compare the enhancement factor and resonance width of RFWM with the theoretical values, the factor w/γ must be determined.

5 Damping Mechanisms

5.1 Landau Damping

In order to find the theoretical enhancement in reflectivity, it is necessary to find the correct damping rate. For ion acoustic waves, the important damping mechanism generally is ion Landau damping. The Landau damping rate decreases exponentially with the ratio $\theta = ZT_e/T_i$. If the temperature is assumed to be approximately equal, so that $\theta = 1$, then the damping rate is approximately

$$\frac{\text{Im}w}{\text{Re}w} = 0.3 \quad (19)$$

This implies that the reflectivity should be enhanced by a factor of about 10, while the width of the resonance peak should be quite broad. For argon,

Figure 5: Mass Dependence

with a resonant frequency of 30.5 MHz, the damping rate should be 10 MHz. This width is much broader than the experimentally measured width.

Landau damping, however, is not the important damping mechanism in this case. The ion-ion collision frequency, which is proportional to the density and inversely proportional to the temperature, is on the order of 10^{10}s^{-1} . The collision frequency is much higher than the acoustic frequency w_{ac} , so Landau damping cannot occur.

5.2 Collisional Narrowing

When the plasma is highly collisional, other damping mechanisms become more important than ion Landau damping. Mostovych and DaSilva, and later Zang and DaSilva, studied the thermal ion acoustic fluctuation spectrum of a collisional plasma, and found that the width of the resonance peaks are much narrower than the predictions of Landau damping.

Mostovych and DaSilva performed their experiments in a dense, low temperature argon plasma with typical values of $n_e = 10^{17}\text{cm}^{-3}$ and $T_e = 2\text{eV}$. They directed a 200 W CO_2 laser beam into the plasma, and observed the scattered signal. The power spectrum of light scattered by a plasma contains information about the density fluctuations in the plasma. Their data showed peaks corresponding to thermal ion fluctuations (See Figure 6). The resonance, however, is much narrower than Landau damping predicts, because of the high collision frequency. The high collision rate reduces the effectiveness of Landau damping, and the plasma becomes dominated by collisional and thermal damping. Their analysis leads to a theoretical resonance width of about 6 MHz for Argon, and about 12 MHz for Nitrogen. Their analysis, however, does not include electron-ion coupling effects, which may be very important in our experiment.

A complete analysis of damping in the collisional regime has been performed by Federici. His analysis includes collisional damping, thermal damping, and viscous damping, and is valid if the following conditions are valid:

$$w_{mn} < \nu_e \quad (20)$$

$$\frac{w_{fp}\nu_e}{w_{pi}^2} \ll 1 \quad (21)$$

$$\frac{\nu_e m_e}{w_{fp} m_i} \ll 1 \quad (22)$$

Figure 6: Thermal Ion Fluctuations : DaSilva

These conditions are all met in our experimental set-up. Assuming a temperature of $T_e = T_i = 2.25$ eV, and a density $n_0 = 9.5 \times 10^{16} \text{cm}^{-3}$, the important parameters are $w_{ac} = 1.92 \times 10^8 \text{sec}^{-1}$, $\nu_{ei} = 2.89 \times 10^{11} \text{sec}^{-1}$, and $w_{pi}^2 = 4.12 \times 10^{21} \text{sec}^{-2}$. The damping rate in this analysis is about $\gamma = 0.1 w_{ac}$. This is much smaller than Landau damping predicts, and fits fairly well with the experimentally measured width. A plot of normalized reflectivity vs frequency is shown in Figure 7.

6 Convection

Although this model predicts the resonance width fairly well, it also predicts a very large enhancement. Since the density increase is proportional to w/γ , this model should increase the density by a factor of 10, and the reflectivity by a factor of 100. However, the experimental enhancement is only about 4. There are many factors that can reduce the signal from the large predicted value. The first important factor is due to convection effects. Although the plasma resonance increases the density modulation significantly, the propagating ion acoustic wave is also moving energy out of the plasma. Since the three beams interact in a 1mm spot, there are only a few grating periods in the region. For the small k grating, with a wavenumber $k_{fp} = 413.74 \text{cm}^{-1}$, the grating wavelength is $\lambda_{fp} = 2\pi/k_{fp} = 0.15 \text{mm}$. This means there are only 5 or 6 grating periods in the region, which is not enough to allow the density modulation to build up to its peak value. In order to determine the density modulation, the differential equation with respect to space must be solved:

$$\left[\frac{\partial^2}{\partial t^2} + \gamma \frac{\partial}{\partial t} + c_s^2 \frac{\partial^2}{\partial x^2} \right] n(x) = \frac{jk}{M} F_{NL} \quad (23)$$

The density profile can no longer be assumed to be constant in the x direction. If the force is assumed to be uniform in the x direction, then convection can reduce the density enhancement at resonance by a factor of about 3. If a more realistic force is assumed, the reduction is even larger. Figure 8 shows the density reduction at resonance, as a function of grating periods in the plasma, for two cases : 1) A flat force and, 2) A force with a triangle profile. A more realistic case would be a gaussian force, which is between these two cases. Clearly, as the size of the plasma increases, convection becomes less and less important. In this case, however, with a size of approximately 1mm,

Enhancement vs. Frequency (Federici)

Figure 7: Density Enhancement : Federici

Convection Effects

Figure 8: Convection Effects at Resonance

convection can reduce the reflectivity enhancement by an order of magnitude. Away from resonance, the effects of convection are not as important. Figure 9 shows how the Lorentzian profile is altered by convection.

7 Temperature Shift

The experimentally measured reflectivity is also reduced from the predicted value because of a temperature shift in the plasma. During the build-up of the laser pulse, the plasma electron temperature gradually begins to rise. The electron temperature rises from 2 eV to about 5 eV. As a result, the plasma resonance is not present for the duration of the laser pulse. In the harmonic oscillator model, the density modulation n_1/n_0 can be solved from the second order differential equation :

$$\left[\frac{\partial^2}{\partial t^2} + \gamma \frac{\partial}{\partial t} - k^2 c_s^2(t) \right] n_1 = \frac{j k}{M} F_{NL} \cos \omega t \quad (24)$$

In this equation, the ion acoustic velocity, $w_{ac} = k c_s$, now varies with time. The density modulation always requires a finite time to build up to its peak value. The ion acoustic velocity, however, changes before the density can reach its peak value. As a result, it can never be enhanced by the factor ω/γ as predicted. The differential equation, with a varying w_{ac} , must be solved in order to determine the density modulation. A plot of the peak density modulation n_1/n_0 versus applied frequency is shown in Figure 10. The changing resonance frequency may also explain the sharpness of the signal pulse shape (see Figure 3), since the resonance is not present for the duration of the incident pulse.

In order to include the effects of convection and the temperature shift, the second order differential equation must be solved with respect to both time and space. Each effect working alone can reduce the density modulation by a factor of about three. If both processes are considered, however, it is not yet known if the two effects will work together. Figure 11 shows a simple attempt to include both convection and temperature shift effects. The differential equation in space was solved first with convection effects only. For a given driver frequency, the reduction by convection was determined for different ion acoustic frequencies. The differential equation in time was then solved with temperature shift effects. As the ion acoustic frequency changed, the driver

Convection

Figure 9: Lorentzian Profile Reduced by Convection

Figure 10: Effects of Temperature Shift

Theory-Temp Shift , Convection
Constant Gamma

Argon Reflectivity

Figure 11: Argon Reflectivity

force was reduced by the appropriate convection factor. At resonance, the driver force was reduced by a factor of three, while far away from resonance, the force was unaffected. Figure 11 shows the reflectivity versus applied driver frequency. The plot shows good agreement with the experimental data for the peak and the resonance width. Far away from resonance, however, the experimental data does not fit quite as well.

This model was also used to analyze the temporal profile of the signal pulse. Figure 12 shows some sample pulse shapes for three different normalized driver frequencies. The location of the peak is determined by the changing temperature of the plasma and the profile of the driving force. In the first figure, the ion wave is initially out of resonance, and then moves into resonance as the plasma temperature increases. As the temperature continues to rise, the wave goes out of resonance, but the driving force is still increasing. Later, the plasma moves back in resonance as the plasma temperature decreases, but the force is not at its maximum. In the second picture, the plasma resonance almost coincides with the peak of the force. The third picture shows another situation where the resonance and the peak of the force do not occur simultaneously. In both the first and third pictures, the peak density modulation is smaller than the second case.

The experimentally observed signal (See Figure 3) does not exhibit the features shown in the above simulation. For example, two peaks were not observed when the applied frequency was tuned away from resonance. The signal pulse shapes were narrower than the incident pulse, and could correspond to the first of the two peaks shown in Figure 12. The signal, however, does not build back up as this simulation predicts.

8 Conclusion

The degenerate four wave mixing experiments clearly show the plasma is in the collisional regime. The importance of the thermal force can be seen by the dominance of the small k grating. Also, the reflectivity scales as n_0^6 , which indicates the thermal force is the main nonlinear mechanism.

In Resonant Four Wave Mixing, the reflectivity shows enhancement at the ion acoustic frequency. When the gas is changed, the resonance frequency shifts because the ion acoustic velocity changes. The resonance frequency varies as $1/\sqrt{M}$, as the theory predicts. From the width of the resonance

Figure 12: Density Squared Profiles

curve, it is clear that collisional damping is the important damping mechanism. The enhancement, however, is not as large as expected, due to the effects of convection and the temperature shift. If these effects could be reduced, by increasing the plasma size and decreasing the laser intensity, then Resonant Four Wave Mixing could be a very accurate diagnostic for the plasma temperature, the ion acoustic velocity, and the damping rate.

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