5

Spectral Properties of Plasma from a Planar Water Microjet Irradiated at Oblique Incidence

5.1 Electromagnetic wave propagation in a homogeneous plasma

Plasma, being a collection of positive and negative charges, responds to external electromagnetic disturbances and modifies the external fields. This forms the core of the discussion about the propagation of an electromagnetic radiation through a plasma. The transmission or reflection of an electromagnetic radiation of frequency ω in a plasma is basically decided by the plasma frequency. If the frequency of the incoming radiation is larger than the plasma frequency, which gives the fundamental timescale on which a plasma responds to an external electromagnetic disturbance, the radiation is transmitted by the plasma. If the frequency of the electromagnetic radiation is less than the plasma frequency, plasma will reflect the electromagnetic radiation. When there is a gradient in the plasma density the plasma frequency will also be varying. At the electron density where the plasma frequency becomes equal to the frequency of the electromagnetic radiation, the wave will be reflected from the plasma. This electron density is named as the *critical density* and it is given by

$$n_{cr} = \frac{\varepsilon_0 m_e \omega^2}{e^2} \qquad (5.1)$$

Let us now consider the propagation of a high frequency electromagnetic radiation through a plasma. The frequency of the electromagnetic radiation is chosen such that it is greater than the plasma frequency, thus allowing propagation through the plasma. The plasma is assumed to be devoid of any large imposed or self-generated magnetic field. The ions are considered to form a stationary neutralizing background. The propagation of a plane wave $\mathbf{E} = \mathbf{E}_0 e^{-i(\omega t - \mathbf{k.r})}$

through the plasma can be analyzed by obtaining the wave equation for the oscillating electric and magnetic fields in the plasma using Farady's and Ampere's laws. Since the current density in the plasma is given by the motion of the electrons under the electric field of the electromagnetic radiation, a linearized force equation for the electron will give the current density. Thus the Helmholtz equations for the electric and magnetic fields of the electromagnetic wave in the plasma are given by

$$\nabla^2\mathbf{E} - \nabla(\nabla.\mathbf{E}) + \frac{\omega^2}{c^2}\varepsilon\mathbf{E} = 0 \tag{5.2}$$

and

$$\nabla^2\mathbf{B} + \frac{\omega^2}{c^2}\varepsilon\mathbf{B} + \frac{1}{\varepsilon}\nabla\varepsilon \times (\nabla\times\mathbf{B}) = 0 \tag{5.3}$$

respectively, where $\varepsilon = 1 - \frac{\omega_{pe}{}^2}{\omega^2}$ is the dielectric function of the plasma. For a homogeneous plasma the electron density will be spatially uniform and hence the dielectric constant of the plasma will be uniform. There will not be any charge accumulation. As a result, for a homogeneous plasma $\nabla\varepsilon = 0$ and $\nabla.\mathbf{E} = 0$. Then the wave equations for the electric and magnetic fields in the plasma will become

$$\nabla^2\mathbf{E} + \left(\frac{\omega^2}{c^2}\right)\varepsilon\mathbf{E} = 0 \tag{5.4}$$

and

$$\nabla^2\mathbf{B} + \left(\frac{\omega^2}{c^2}\right)\varepsilon\mathbf{B} = 0 \tag{5.5}$$

respectively. The dispersion relation for the electromagnetic wave in the *homogeneous* plasma is given as:

$$\left(\frac{\omega^2}{c^2}\right)\varepsilon = k^2 \qquad \text{or} \qquad \omega^2 = \omega_{pe}{}^2 + k^2c^2 \tag{5.6}$$

5.2 Electromagnetic wave propagation in an inhomogeneous plasma

In an *inhomogeneous* plasma the electron density is no longer uniform and hence the full wave equations have to be solved to get the electric and magnetic fields in the plasma. Under such a situation the waves will no longer be plane waves once they propagate through the plasma. Depending on the nature of polarization and angle of incidence on the plasma the fields will evolve to different solutions. The nature of the plasma density profile will be reflected on the nature of the solutions for the electric and magnetic fields that propagate through the inhomogeneous plasma. By assuming the plasma density profile, the propagation equation can be solved to get the field distribution.

5.2.1 Oblique incidence of a plane electromagnetic wave

Consider an electromagnetic wave whose propagation vector is at an angle to the electron density gradient in the plasma. For the analysis we can consider a plane electromagnetic wave incident on a plasma slab of electron density $n_e(z)$. The vacuum-plasma interface is taken as z = 0, where the angle of incidence θ is defined as the angle between the propagation vector and the direction of the density gradient ($\hat{z}$). The position of the electron critical density layer from the vacuum-plasma interface is denoted by L. The plane of incidence of the electromagnetic wave is defined by the vectors ∇n and **k**. For the analysis let it be the y–z plane, with the density gradient along the z direction. Under this labeling there is no variation along the x direction (i.e. $k_x = 0$ and $\frac{\partial}{\partial x} = 0$). At the vacuum-plasma interface (z = 0) the y and z components of the propagation vector are given as $k_y = \left(\frac{\omega}{c}\right)\sin\theta$ and $k_z = \left(\frac{\omega}{c}\right)\cos\theta$. The propagation of the electromagnetic wave in the plasma under oblique incidence will dependent on whether the electric field vector E of the incident radiation is in or out of the above defined plane of incidence [Kruer, W.L.].

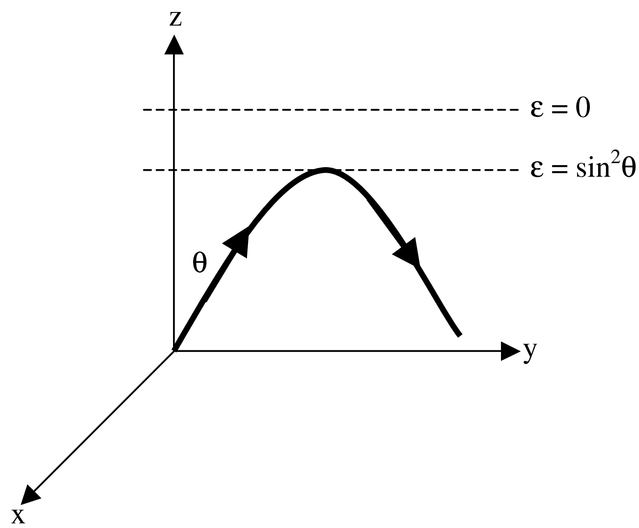

Figure 5.1: Light ray obliquely incident on an inhomogeneous plasma slab

S-polarized electromagnetic wave

If the electric field vector is normal to the plane of incidence, the electromagnetic wave is called *s-polarized*. If we take $\mathbf{E} = \mathrm{E}_x \hat{x}$, following which the propagation vector and the plasma density gradient are taken at the y-z plane, then the wave equation of the electric field vector will become:

$$\frac{\partial^2 E_x}{\partial y^2} + \frac{\partial^2 E_x}{\partial z^2} + \frac{\omega^2}{c^2}\varepsilon(z)E_x = 0 \tag{5.7}$$

under the assumption that no variation occurs along the x direction. Since the dielectric function of the plasma is a function of z alone, k_y should be conserved. Hence we get

$$E_x = E(z)\exp\left(\frac{i\omega y \sin\theta}{c}\right) \tag{5.8}$$

E(z) will become zero or the wave will be reflected when $\varepsilon(z) = \sin^2\theta$ as it is clear from the wave equation with the substitution of E_x:

$$\frac{d^2 E(z)}{dz^2} + \frac{\omega^2}{c^2}\left(\varepsilon(z) - \sin^2\theta\right)E(z) = 0 \tag{5.9}$$

Reflection occurs at $\omega_{pe} = \omega \cos\theta$, which implies that an obliquely incident electromagnetic wave will reflect at a lower density than the critical density for that wavelength, i.e., at an electron density of $n_e = n_{cr}\cos^2\theta$. For a plasma with a linear density profile the wave reflects at z = L$\cos^2\theta$.

P-polarized electromagnetic wave

When the electric field vector of the electromagnetic radiation lies parallel to the plane of incidence it is known as *p-polarized*. Under the coordinate consideration of figure 5.1, the electric field can be written as $(E = E_y\hat{y} + E_z\hat{z})$. It is obvious that the electric field has a component in the z direction, which is the direction of free electron density gradient assumed. This leads to a non-zero product $(\mathbf{E}.\vec{\nabla}n_e \neq 0)$. i.e. $\left(\mathbf{E}.\vec{\nabla}n_e = E_z\frac{\partial n_e}{\partial z}\right)$. This component of the electric field of the obliquely incident electromagnetic radiation will make the electrons oscillate along the direction of the electron density gradient. Since this oscillation of electrons generates fluctuations in charge density, which can be resonantly enhanced by the plasma, the wave is no longer purely electromagnetic. Part of the energy of the incident electromagnetic wave is transferred to an electrostatic oscillation, the electron plasma wave (Langmuir wave), a phenomenon named as *Resonance Absorption (RA)*.

Consider a plane electromagnetic wave incident at an angle θ onto an inhomogeneous plasma slab with density $n_e(z)$ as shown in figure 5.1. Poisson's equation gives $\vec{\nabla}.(\varepsilon\mathbf{E}) = 0$, which readily gives the divergence of the electric field of the incident electromagnetic radiation in the plasma as:

$$\vec{\nabla}.\mathbf{E} = -\frac{\mathbf{E}.\vec{\nabla}\varepsilon}{\varepsilon} = -\frac{1}{\varepsilon}\left(\mathrm{E}_z\frac{\partial\varepsilon}{\partial z}\right) \tag{5.10}$$

with ε as the dielectric function of the plasma. The electromagnetic field that satisfies this condition induces a charge separation in the plasma. A non-vanishing derivative of the dielectric function exists only when there is a spatial electron density gradient that reflects in the plasma frequency. Resonant response will be present when ε=0, i.e., when $\omega_{pe}=\omega$.

Physically, resonance absorption can be explained by considering electrons oscillating between regions of differing density. This directly creates a charge density fluctuation, $\delta n = n(\mathbf{x}+\mathbf{x}_{osc}) - n(\mathbf{x}) \cong \mathbf{x}_{osc}.\vec{\nabla} n_e$. The amplitude of oscillation of an electron in the electric field of the electromagnetic radiation is given by $\left(\mathbf{x}_{osc} = e\mathbf{E}/m\omega^2\right)$. When $\omega_{pe}=\omega$, this imposed charge density fluctuation is just at the frequency where the plasma responds resonantly. Hence an electron plasma wave is excited at ε=0 (i.e. at the critical density), when an electromagnetic wave is obliquely incident on an inhomogeneous plasma. An obliquely incident electromagnetic radiation will be reflected from an inhomogeneous plasma at an electron density less than the critical electron density at the incident frequency. For a *p*-polarized electromagnetic wave the field will tunnel into the critical density region and excite the plasma wave resonance, even when it is reflected at a density less than the critical density.

To obtain the electric field along the direction of the electron density gradient, the first step is to get the magnetic field of the *p*-polarized electromagnetic wave. At each position of the density gradient, the magnetic field of the electromagnetic radiation is $\mathbf{B} = \hat{x}B_x$, which will be normal to both the propagation vector **k** and the direction of electron density gradient, which is taken as the z direction in the present treatment. Considering the conservation of the y component of the propagation vector, the magnetic field of the electromagnetic wave can be written as:

$$\mathbf{B} = \hat{x}B(z)\exp\left(-i\omega t + \frac{i\omega y \sin\theta}{c}\right) \tag{5.11}$$

Applying Ampere's law to the magnetic field as $\vec{\nabla}\times\mathbf{B}=-\frac{i\omega}{c}\varepsilon\mathbf{E}$ and equating the z components, the amplitude of the electric field along the z direction (i.e. the electrostatic field along the direction of electron density gradient, which will have a resonance behaviour near the critical density layer) is given as:

$$E_z=\frac{\sin\theta}{\varepsilon(z)}B(z) \tag{5.12}$$

Since E_z is strongly peaked at the critical density, the resonantly driven field is approximated as ${E_d}/{\varepsilon(z)}$, with E_d evaluated at the resonance point. Physically E_d is the *field driving the resonance*, i.e. the component of the electromagnetic wave that oscillates electrons along the density gradient at the critical density. To evaluate E_d, we need the magnetic field at the critical density. For this we need to calculate the magnetic field along the direction of the electron density gradient. This can be done by considering the wave propagation in an inhomogeneous plasma. Assuming a linear electron density profile, the value of the magnetic field B(z) at the critical density layer z=L can be obtained by multiplying the value of the magnetic field at the turning point of the obliquely incident *p*-polarized electromagnetic radiation ($B(z=L\cos^2\theta)$) by an exponential decay from the turning point to the critical density. The value of the magnetic field of the electromagnetic radiation at the turning point can be estimated using the Airy function solution for an s-polarized wave. Hence $B(z=L\cos^2\theta)\approx 0.9E_{FS}\left({c}/{\omega L}\right)^{1/6}$. Here E_{FS} is the value of the electric field of the light wave in free space. The decay of the field as it is penetrated beyond the turning point is estimated by $e^{-\beta}$, where

$$\beta=\int_{L\cos^2\theta}^{L}\frac{1}{c}\sqrt{\omega_{pe}^2-\omega^2\cos^2\theta}\,dz \tag{5.13}$$

For a linear density profile β is $\left(2\omega L / 3c\right)\sin^3\theta$. Hence the magnetic field at the critical density layer of an obliquely incident *p*-polarized electromagnetic radiation is given as:

$$B(z=L) \cong 0.9 E_{FS} \left(c / \omega L\right)^{1/6} \exp\left(-\frac{2\omega L \sin^3\theta}{3c}\right) \tag{5.14}$$

Hence the component of electric field that oscillates the electrons along the density gradient is given as ($E(z)\varepsilon(z) = B(z)\sin\theta$):

$$E_d \cong 0.9 E_{FS} \left(c / \omega L\right)^{1/6} \sin\theta \exp\left(-\frac{2\omega L \sin^3\theta}{3c}\right) \tag{5.15}$$

Which can be rewritten in terms of a parameter τ (defined as $\tau = \left(\omega L / c\right)^{1/3} \sin\theta$) as:

$$E_d = \frac{E_{FS}}{\sqrt{2\pi\left(\omega L / c\right)}} \varphi(\tau) \tag{5.16}$$

with $\varphi(\tau) = 2.3\tau \exp\left(-2\tau^3 / 3\right)$. Hence the driver field vanishes as $\tau \to 0$ and becomes very small for large τ. For τ to be large θ should be large, and under this condition the incident wave has to tunnel through a large distance to reach the critical density layer. Hence there is an optimum angle of incidence for a *p*-polarized electromagnetic radiation to resonantly excite the plasma wave upon oblique incidence to an inhomogenous plasma. This is given approximately by the condition, $\left(\omega L / c\right)^{1/3} \sin\theta \approx 0.8$.

Resonance energy absorption

The resonantly driven electric field is $E_z = E_d / \varepsilon(z)$. If we consider a damping of the wave with a damping frequency ν, which can represent dissipation by electron-ion collisions, linear or non-linear wave-particle interactions, or propagation of the wave out of the resonant region, the dielectric function of the plasma can be written as $\varepsilon(z) = 1 - \left(\omega_{pe}^2(z) \big/ \omega(\omega + i\nu) \right)$. The maximum value of the electric field along the electron density gradient direction is proportional to ν^{-1} and the width of the resonance of E_z near the critical density layer is proportional to ν. For a linear density profile the absorbed energy flux is given as

$$I_{ab} \cong \frac{\omega L E_d^2}{8} \tag{5.17}$$

5.2.2 Normal incidence of a plane electromagnetic wave

Exact solutions can be obtained for a plane electromagnetic wave normally incident on a plasma slab with a linear variation in density. Consider the electric field vector to be in the x direction and let $E_x = E$ (*s*-polarized wave according to our conventions of figure 5.1). Considering the variations only in the direction of propagation of the plane electromagnetic wave (z direction), in cartesian coordinates, the wave equation for the electric field becomes,

$$\frac{d^2E}{dz^2} + \left(\frac{\omega^2}{c^2} \right) \varepsilon(\omega, z) E = 0 \tag{5.18}$$

Assuming that plasma density (electron density) is a linear function of position along the direction of propagation of the electromagnetic radiation under the condition of normal incidence to the plasma slab, $n = n_{cr} z \big/ L$, the wave equation will become:

$$\frac{d^2E}{dz^2} + \left(\frac{\omega^2}{c^2} \right) \left(1 - z \big/ L \right) E = 0 \tag{5.19}$$

and assuming a change of variable as $\eta = \left(\frac{\omega^2}{c^2 L} \right)^{1/3} (z - L)$ the wave equation will become a differential equation given by $\frac{d^2E}{d\eta^2} - \eta E = 0$ with a general solution in terms of the Airy functions as $E(\eta) = \alpha A_i(\eta) + \beta B_i(\eta)$, where the constants α and β are determined from the boundary conditions. Hence,

$$E(\eta) = 2\sqrt{\pi}\left(\frac{\omega L}{c}\right)^{1/6} E_{FS} e^{i\varphi} A_i(\eta) \tag{5.20}$$

with E_{FS} as the free space electric field of the incident electromagnetic radiation. The amplitude of the electric field reaches a maximum at the cut off layer (ε=0, n_e= n_{cr}) due to the constructive interference between the incident wave and the wave reflected from the plasma critical density layer. This maximum value of the electric field is given by

$$\left|\frac{E_{\max}}{E_{FS}}\right|^2 \cong 3.6\left(\frac{\omega L}{c}\right)^{1/3} \tag{5.21}$$

The magnetic field of the electromagnetic wave in an inhomogeneous plasma with a density gradient along the direction of propagation of the incident plane wave is given as

$$B(\eta) = -i2\sqrt{\pi}\left(\frac{c}{\omega L}\right)^{1/6} E_{FS} e^{i\varphi} A'_i(\eta) \tag{5.22}$$

where the prime denotes derivative with respect to η. At the reflection point,

$$\left|B(\eta = 0)\right| \approx 0.92\left(c/\omega L\right)^{1/6} E_{FS} \tag{5.23}$$

In the case of normal incidence of a plane electromagnetic wave on inhomogeneous plasma, the wave propagation equations inside the plasma are not dependent on whether the electromagnetic wave is *s* or *p* polarized. In chapter four the situation of normal incidence (of a *p*-polarized laser pulse) to a planar water jet was considered.

5.3 Mechanisms of plasma wave damping

As discussed in the previous section, in resonance absorption, large amplitude electrostatic waves are produced due to the field resonance at the critical layer. Transfer of energy from this electrostatic wave to the surrounding plasma occurs through damping [Kruer, W. L.]. There are collisional and non-collisional ways of damping the energy of resonantly excited plasma waves, leading to the generation of 'hot' (non thermal) electrons. Collisions between the electron wave and the ions of the plasma lead to the transfer of energy from the electrostatic wave to the ions. This is similar to the energy transfer from an electromagnetic radiation to the ions via electrons that undergo quiver motion in the incident laser field. The coherent motion of the electrons, as the plasma wave, will be changed to a random motion via this collision with the surrounding ions.

Even in the absence of collisions the electrostatic waves in a plasma can be damped. *Landau damping* is one such mechanism in which particles surrounding the electrostatic waves exchange energy with the wave depending on their velocity. The velocity of the particles in the plasma with respect to the phase velocity of the plasma wave determines the energy exchange. Those particles whose velocities are not resonant to the phase velocity of the plasma will not gain or lose energy. Those particles with velocities comparable to the phase velocity of the wave will see a nearly constant electric field from the plasma wave, and will thus get accelerated or decelerated. For particles with a velocity distribution f(ν), the rate at which the particle gains or loses energy from the plasma wave is given by

$$\frac{d\Sigma}{dt} = -\frac{\pi q^2 E_w{}^2}{2m|k_w|}\frac{\omega_w}{k_w}\left(\frac{\partial f}{\partial v}\right)_{\omega_w/k_w} \tag{5.24}$$

where the last term represents the slope of the velocity distribution of the particles at the phase velocity of the wave. The subscript 'w' in the above expression represents the parameters of the plasma wave as against those of the electromagnetic wave. This shows that particles with a velocity slightly less than the plasma wave phase velocity will gain energy from the wave, and those having slightly greater velocity will lose energy to the wave [Kruer, W. L.]. Landau damping addresses wave-particle interaction in the linear regime.

If the amplitude of the plasma wave is large, even extremely slow electrons can be accelerated in the electric field bringing their velocities in resonance with the phase velocity of the wave. This indicates that under such conditions, any electron moving in the electric field of the plasma wave will be resonant with it. This leads to a strong, nonlinear damping of the plasma wave as many electrons are efficiently accelerated to resonance with the wave thus extracting energy from the wave. A maximum number of electrons is brought to resonance at the peak of the wave and hence maximum damping will occur there. As a result, different parts of the wave will have different velocities depending on the field amplitude at those positions. This non-uniformity in the velocity will lead to the breaking of the plasma wave. The *wave breaking* condition can be written as:

$$\frac{eE_w k_w}{m\omega_w^2} \geq 1 \tag{5.25}$$

5.4 Polarization dependent second harmonic emission from plasma

When an intense femtosecond laser pulse interacts with solid density matter (solid or liquid) a very thin layer of high temperature, high-density plasma is generated on the surface of the material. During the laser pulse, the plasma density drops from nearly solid density essentially to zero in a distance much shorter than the wavelength of the laser light. Oblique reflection of *p*-polarized electromagnetic wave from an overdense, inhomogeneous plasma will be accompanied by a second harmonic (SH) emission [Eidmann & Sigel]. When resonance absorption takes place in a plasma, SH is generated by an electron plasma wave at the critical density layer. The frequency of the SH under such a situation will be exactly twice that of the fundamental frequency, with the signature of the Doppler motion of the critical layer. SH in the specularly reflected direction and nearly twice the frequency of the fundamental are characteristic of resonant absorption processes [Hansen *et al.*]. When the characteristic size of the underdense plasma is small $(L/\lambda \leq 10)$ the primary consideration is the energy absorption near the critical density surface. If the length of the under dense plasma is large $(L/\lambda \geq 100)$ phenomena like inverse Bremsstrahlung absorption, Brillouin and Raman scattering will be significant. Electron plasma waves generated by resonance absorption account for the SH emission. The formation of SH from plasma can be explained in a simple manner as given below.

When *p*-polarized light is incident on an inhomogeneous plasma slab, electron plasma waves are excited at the critical density layer via resonance absorption mechanism. The density oscillation of the excited plasma wave non-linearly couples with the oscillatory velocity of the plasma electrons resulting in an ac electronic current in the plasma that eventually radiates the SH of the incident light. By considering perturbation theory, the current density at 2ω can be written as:

$$\vec{J}_{2\omega} = e\left(n^{(0)}\vec{v}_{2\omega}^{(2)} + n_{\omega}^{(1)}\vec{v}_{\omega}^{(1)} + \ldots\ldots\ldots\ldots\ldots\right) \tag{5.26}$$

where $\vec{v}_{\omega}^{(1)}$ and $\vec{v}_{2\omega}^{(2)}$ are the first and second order electron velocity perturbations at ω and 2ω respectively, $n_{\omega}^{(1)}$ is the first order perturbation of the electron density, and $n^{(0)}$ is the unperturbed density profile of the plasma [Linde *et al.*]. By combining Maxwells equations, the continuity equation and the plasma equation of motion, the SH current density is found to be related to the local electric field in the plasma as

$$\vec{J}_{2\omega} = \frac{n^{(0)}e^3}{4m_e^2\omega^3}\vec{\nabla}\left(\vec{E}_{loc}.\vec{E}_{loc}\right) + \frac{e^3}{m_e^2\omega^3}\left(\frac{\vec{\nabla}n^{(0)}.\vec{E}_{loc}}{1-\omega_{pe}^2/\omega^2}\right)\vec{E}_{loc} \tag{5.27}$$

where $\vec{E}_{loc}$ is the local electric field oscillating at the laser frequency, and $\vec{\nabla}n^{(0)}$ is the longitudinal electron density gradient [Gizzi *et al.*]. Solving the wave equation with $\vec{J}_{2\omega}$ as a source term will show that SH emission will occur from the plasma. Thus, the coherently driven nonlinear current acts as the dominant source for SH from the plasma. Conservation of the wave vector component parallel to the surface leads to the emission of the SH in the form of a coherent beam collinear with the specularly reflected fundamental beam [Gizzi *et al.*],[Linde *et al.*]. Since the electron density and velocity perturbations referred in equation 5.26 are directly driven by the fundamental laser field at ω, the SH spectrum is directly related to the fundamental spectrum through the current density at 2ω. Plasma expansion will introduce Doppler shift to the SH frequencies, which in the static model, is centered at 2ω [Linde *et al.*]. A red shift of the SH is possible due to the phase change of the nonlinear current in the plasma during its evolution. The major component of the red shift of the SH arises from the second order current density shown above. The red shift of the SH can be attributed to the changes of the phase of the nonlinear current during the evolution of the plasma. In the initial stages of the plasma the light is essentially interacting with a supercritical electron density, and the induced electron density oscillations are in phase with the laser field. Once the plasma begins to expand, the laser will be interacting with critical density or sub-critical density electrons. The induced electron density oscillations are out of phase with the field in this condition. Thus the nonlinear current density will have an increasing phase lag with the laser field as time progresses. This phase lag will be reflected as a red shift in the SH spectrum since $\Delta\omega = -\frac{d\phi}{dt}$. Since this time varying phase is for the second order current density, the first order optical properties, i.e. responses in the fundamental frequency, will be unaffected. Thus the red shifted SH is consistent with a blue shifted fundamental [Linde *et al.*].

The blue shift of the fundamental light frequency upon reflection from the inhomogeneous plasma can be attributed to the Doppler shift induced in the plasma heating pulse (in femtosecond laser-matter interaction, the part of the laser pulse after plasma creation) by the motion of mass from the target surface [Milchberg & Freeman]. The heated target material will move away from the original interface during the evolution of the pulse. The expanding interface acts as a moving mirror. Thus the reflected spectrum of the laser pulse is a convolution of the laser pulse history and a time-dependent frequency shift set by a time-dependent expansion speed. The spectral function of the reflected light depends on the spectral function of the incident light as follows:

$$S_R(\omega) \propto \int_{-\infty}^{\infty} I(t) S_i(\omega - \Delta\omega(t)) R(t) dt \tag{5.28}$$

where $S_i(\omega)$ is the spectral function of the incident light, I(t) is the laser pulse intensity, and R(t) is the reflectivity of the material. The Doppler shift in frequency is given as:

$$\Delta\omega(t) = \left(\frac{2\omega}{c}\right)(\cos\theta) v(t) \tag{5.29}$$

with θ being the angle of incidence and $v(t)$ the spatial average velocity, along the transverse to the expansion direction, of the reflection surface in the beam focus. Thus the reflected spectrum depends on the change of $v(t)$ during the pulse. At the typical plasma density resulting from femtosecond laser interaction, the plasma pressure exceeds the light pressure, and hence the expansion is solely determined by the plasma pressure. The spectrum of the reflected light represents the time-integrated effect of an expansion speed averaged over a velocity profile and transverse direction of the laser pulse. Thus the temperature obtained from the reflected pulse spectrum is a time average over these space averaged expansion speeds. The spectral shift is given by

$$\Delta\lambda = 2\left(\frac{v_{\exp}}{c}\right)(\cos\theta)\lambda \tag{5.30}$$

where $v_{\exp}$ is the average expansion velocity, c is the velocity of light in free space, θ the angle of incidence, and λ is the central wavelength of the fundamental input laser pulse. The assumption of plasma as a fluid will give an expansion speed of

$$v_{\exp} = \frac{2}{\gamma - 1} c_s \tag{5.31}$$

with the consideration that the initial sharp boundary of a semi-infinite fluid will expand with this speed. Here γ is the ratio of the specific heats of the fluid, which can be chosen to be 1.7 for the plasma of interest pertaining to this experiment. 'c_s' is the sound speed of the fluid. The plasma sound speed can be given as

$$c_s = \left({Zk_B T_e}/{m_i} \right)^{1/2} \qquad (5.32)$$

where T_e is the electron temperature, k_B is the Boltzman constant, Z is the average ion charge state and m_i is the ion mass. Thus from the spectral blue shift of the fundamental frequency reflected from the expanding plasma, the plasma electron temperature can be inferred [Milchberg & Freeman].

5.5 Experimental results and discussion

We investigated the effect of input laser pulse polarization on the emission from plasma created in a water jet in atmospheric pressure, when irradiated at an oblique angle of incidence. This study essentially addresses the effective coupling of laser to plasma formed on a liquid surface. The previous studies were on metallic, glass or thin plastic film targets [Engers *et al.*], [Linde *et al.*], [Gizzi *et al.*], [Sandhu *et al.*]. In our experiment, laser pulses centered at 797nm have been focused to a planar water jet at an angle of 45^0 using a plano-convex lens of 20cm focal length. The water jet thickness is 250μm and the input laser intensity is 4.2×10^{15} W/cm^2. The polarization of the incident laser pulse is varied from *s* to *p* by using an achromatic half wave plate before the focusing lens. The red shifted second harmonic signal is observed at 406nm with increasing intensity as the input nears *p*-polarization. The SH emission was seen only at the specular reflection direction. The observed SH emission at 406nm is approximately 8nm red shifted from the expected SH position of the excitation wavelength at 797nm. The incoherent white light emitted along with the SH also is polarization dependent, as it peaks when the input is *p*-polarized. However the polarization dependence of SH is found to be sharper than that of the incoherent white light. For instance, the SH signal is enhanced about 3.5 times for the *p*-polarized input laser pulse compared to that for the *s*-polarized input, where the corresponding enhancement of the incoherent emission (450 – 600 nm) is only about 1.75 times. Both the SH and incoherent emissions start increasing from a half wave plate angle of $\pm 30^0$ (where the input polarization is between *s* and *p*) to the completely *p*-polarized state (angle = 0^0). Figures 5.2a and 5.2b show the SH emission from the plasma for different input laser polarizations. Near the *p*-polarization a small dip in the SH signal can be seen though it is not so prominent in comparison to previous reports by Gizzi et al., where the input laser intensity was $5x10^{17}$ W/cm^2.

The maximum SH signal is observed at an angle of $\pm 5^0$ from the fully *p*-polarized state, unlike Gizzi's observation in which it is near $\pm 20^0$. The input intensity in our studies is two orders of magnitude less than that of Gizzi et al. Therefore resonance absorption will be relatively less in our case so that an enhancement happens in the SH emission when more *p*-polarization is present in the input laser pulse. The dip in SH near *p*-polarization can be attributed to plasma wave breaking, even though the dip is not very prominent. Figure 5.3 gives the strength of incoherent emission in the visible region detected simultaneously with the SH emission in the specular reflection direction. The incoherent emission also is found to peak for *p*-polarized input.

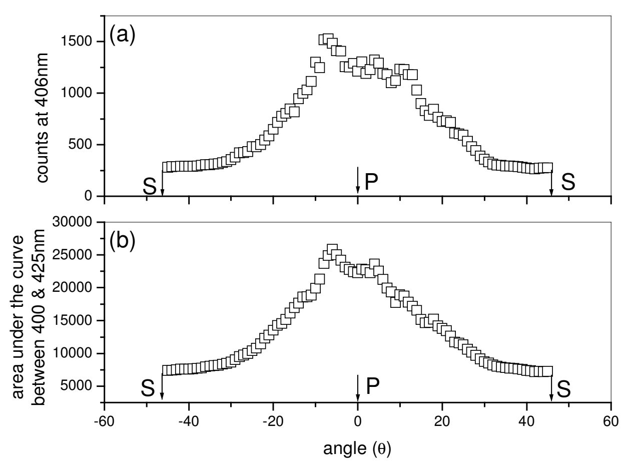

Figure 5.2: (a) Intensity of SH emission at 406nm plotted as a function of the input laser polarization. 0^0 corresponds to *p*-polarized state of the input laser. (b) Area under the curve measured for emission between 400 and 425 nm.

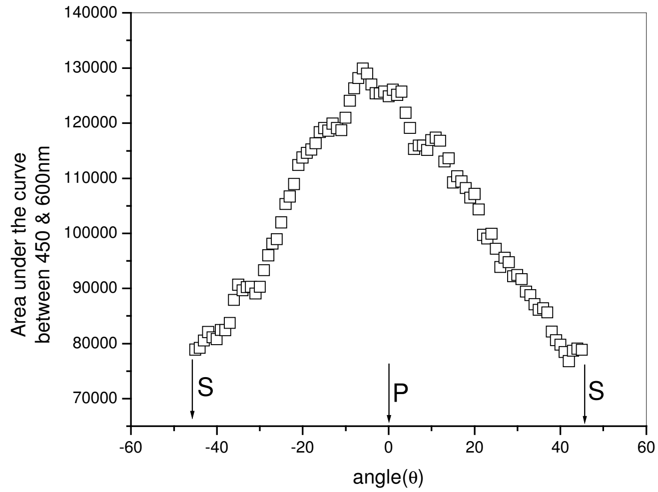

Figure 5.3: Area under the curve for emission between 450 nm and 600 nm (incoherent white light emission) observed in the specular reflection direction.

Both the incoherent emission and SH emission are plotted together in fig. 5.4. As the input approaches *p*-polarization, resonance absorption enhances the coupling of laser intensity to the plasma and the yield of SH is enhanced compared to that of incoherent emission. This can be attributed to the fact that the coherent SH process will be maximized as the plasma waves undergo resonance. The absence of an equal enhancement in the incoherent emission suggests that the plasma waves are still sustained under the resonance condition in the intensity regime of these experiments. The waves are either intact or can be considered to suffer only a weak breaking. The enhancement in incoherent radiation observed near *p*-polarization can be attributed to the increase in laser energy coupling to the electrons imparting them more energy, which eventually results in an increased emission.

Figure 5.5 gives the Doppler shifted fundamental laser pulse, reflected from the plasma. The Doppler blueshift of 4nm suffered by the fundamental laser pulse reflected from the plasma for the oblique incidence of 45^0 suggests an expansion velocity of $2.66 \mathrm{x} 10^5$m/s, according to equation 5.30. By considering

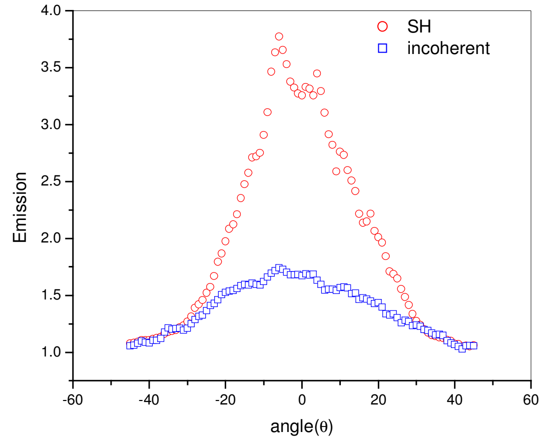

Figure 5.4: Plot of SH emission and incoherent emission. The emission intensities are normalized to a value of 1 for *s*-polarized input.

equations 5.31 and 5.32 and substituting for the expansion velocity the electron temperature can be obtained. The plasma of water in the present experiment contains H^+ ions and O^+ ions as seen from the recombination lines. Hence assuming that the expansion velocity calculated above results from contributions of both of these ions, the average electron temperature can be estimated as 130eV.

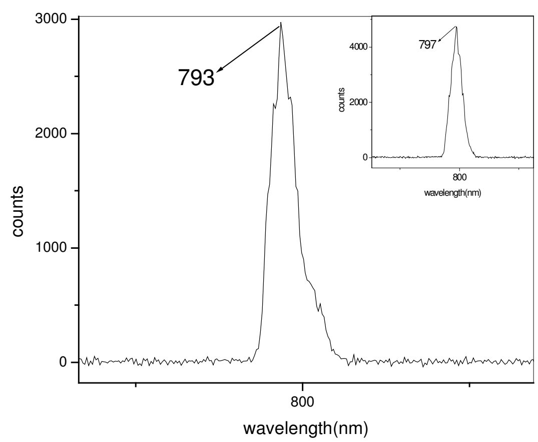

Figure 5.5: Doppler shifted fundamental frequency. A blue shift of 4nm is observed in the fundamental that is reflected at 45^0 from the plasma. Here the input is *p*-polarized. Inset shows the spectrum of the original laser pulse.

Figure 5.6 shows the red shift of the SH which can be attributed to the time-dependent phase change of the second order current density as described in section 5.4.

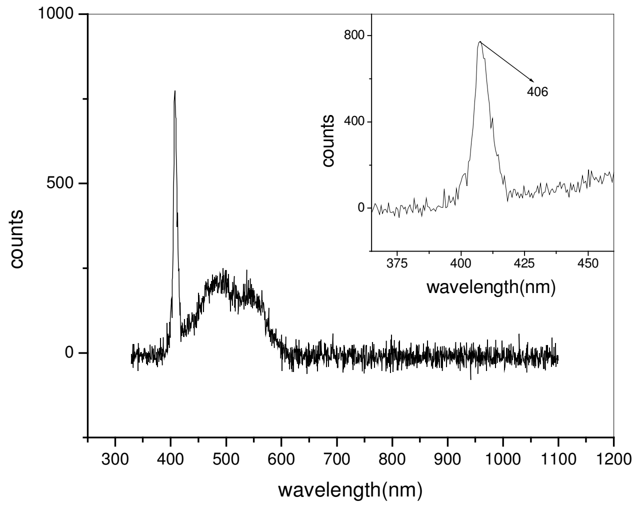

Figure 5.6: SH emission observed at the 45^0 specular reflection direction. An expanded SH is shown in the inset. The peak is at 406nm for this *p*-polarized incidence.

In conclusion, we have investigated laser energy coupling to inhomogeneous plasma under oblique incidence, using 100 femtosecond laser pulses to irradiate a thin planar waterjet. The coupling arising from resonance absorption of the laser photons by the plasma waves has been observed. The coupling is maximized as the input laser pulse approaches *p*-polarization. Detailed polarization dependence studies are done by varying the input polarization from *s* to *p* in steps of 2^0. From a simultaneous detection of SH and incoherent visible radiation, it is shown that SH has more enhancement near the *p*-polarization than incoherent emission. This suggests that at the investigated input laser intensity regime, there is no significant plasma wave breaking. Therefore the coherent SH emission process is more favoured compared to the incoherent emission process. Doppler shift of the fundamental suggests that the expansion velocity is approximately 3×10^5 m/s.