Hot electron generation by highly efficient absorption of high intensity femtosecond laser light in plasma generated on sub-λ gratings

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Abstract. We report near total absorption of light in the interaction of intense, p-polarized ultrashort laser pulse with solid density plasma formed on a gold coated glass sub-λ grating structure aided by surface plasmon resonance (SPR). We measure absorption over a wide intensity range ($2 \times 10^{12} W cm^{-2} - 2 \times 10^{15} W cm^{-2}$). We compare the data with those obtained from highly polished ($\lambda/10$) Au mirror target under identical conditions. The hard X-ray spectrum shows a hotter electron component under the SPR condition.

1. Introduction

Ultraintense, ultrashort laser produced plasmas are sources of hot suprathermal electrons and their signature bremsstrahlung hard x-ray radiation. Such copious fluxes of high energy electrons are of extreme importance to many areas of basic science as well as technology [1]. The need to explore novel ways to optimize the hot electron generation from laser produced plasmas as well as finding ways to control it cannot be overemphasized. Novel target structures are designed and new experimental configurations have been tried to couple more and more light to the ensuing plasma which in effect yield hotter electrons and harder x-rays [2, 3, 4].

Here we use a simple one dimensional periodic system and experimentally pinpoint the window where the incident light is almost completely absorbed resulting in very hot electrons at moderately low light intensity. We believe this is the first systematic study of reflectivity of high intensity ultrashort pulsed light from sub-λ gratings. We model our data using C-method [5, 6] along with Helmholtz solver [7].

2. Experiment

The experiments are performed using a Ti: Sapphire chirped pulse amplified laser (Thales Laser, Alpha 10) emitting 55 fs pulses centered at 800 nm wavelength at 10 Hz repetition rate. The laser is focussed at oblique incidence with a f/20 lens on targets housed in a vacuum chamber at $10^{-4}$ torr. The maximum pulse energy used in the present set of experiments, gives a peak intensity of about $3 \times 10^{15} W cm^2$ at a 60 μm focal spot. A thin half wave plate in the beam path selects the polarization state of light field. The intensity of incident light is controlled by using high contrast thin film polarizer and half wave plate combinations. The target is constantly translated...
in the focal plane in order to avoid multiple laser hits at the same spot. The reflected signal is collected using integrating spheres and calibrated photodiodes. The hard x-ray bremsstrahlung (25 keV - 250 KeV) emission under high intensity laser irradiation is measured using a properly calibrated NaI(Tl) scintillating detector looking normally into the target. Background noise and pile up problems are eliminated by using (i) time gated data acquisition (30 µs collection time window opened in synchronization with the incident laser pulse) and (ii) keeping hard x-ray count rate below one per second, i.e. 0.1 per laser shot by adjusting the solid angle subtended into the detector.

In our experiments we used two types of targets. We used triangular blazed gratings (period, \(d = 555 \text{ nm}\); groove depth, \(h = 158 \text{ nm}\), blaze angle 17.45°) with gold coating on glass substrate (Fig.1) and compared all the data against polished (roughness \(\lambda/10\)) gold coated glass targets. The thickness of gold layer in both the cases is many times greater than the optical skin depth, \(\delta_{\text{s}} \approx c/\omega_{\text{p}} = 21.8 \text{ nm}\) in our case (\(\lambda = 800 \text{ nm}\)) implying that in all our experiments the glass background does not have any significant role to play.

3. Results and Discussion

The dielectric constant for Au (\(\epsilon_m\)) at \(\lambda = 800 \text{ nm}\) is extracted from the s and p reflectivity vs. angle of incidence data by using Fresnel reflectivity formulae for an absorptive medium with a complex refractive index [8]. For Au metal vacuum interface we find, \(\epsilon_m = \epsilon_m' + i\epsilon_m'' = -33.26 + i14.52\), which is reasonably close to the values found in literature [9]. The Drude model prediction, \(\epsilon_m' = 1 - \omega_p^2/\omega^2 = -32.81\) matches reasonably with the value we obtain experimentally implying that at \(\lambda = 800 \text{ nm}\) there is no interband absorption in metallic Au. In our experiments we kept the grating wave vector aligned in the plane of incidence of light. A sub-\(\lambda (\lambda/d > 1)\) grating permits only two diffraction orders, i.e. the specularly reflected one \(m = 0\) and the \(m = -1^{\text{th}}\) order [10]. Low intensity p polarised light shows a sharp dip in specular reflectivity with the minimum at an angle \(\theta_{\text{sp}} = 23^0\). At this angle almost all the light (\(\sim 98.5\%\)) that is incident is completely absorbed (Fig.2). For p polarisation \(\theta_{\text{sp}}\) is the angle at which the phase matching condition is satisfied and it corresponds to a situation where the incident electromagnetic radiation efficiently excites surface charge density oscillations leading to very strong excitation of surface plasmon, i.e. a condition known as surface plasmon resonance(SPR). The phase matching condition for a low amplitude sinusoidal Au grating and vacuum interface is, \(k'_{S\text{p}} = k_0 \sin \theta + 2\pi n/d\), where \(k'_{S\text{p}} = k_0, [(a + \sqrt{a^2 + b^2})/2]^{1/2}\), \(a = [\epsilon_m' + 2\epsilon_m'']/[(1 + \epsilon_m' + \epsilon_m'')^2 + \epsilon_m'']\) and \(b = [2\epsilon_m'/(1 + \epsilon_m' + \epsilon_m'')]\). With \(\epsilon_m = \epsilon_m' + i\epsilon_m'' = -33.26 + i14.52\), \(d = 555 \text{ nm}\) and \(\lambda = 800 \text{ nm}\) the SPR angle comes out to be \(\theta_{\text{sp}} = 26.56^0\) which
is a close match to the experimentally observed angle considering the fact that we have used triangular gratings for our experiments. The coordinate transformation method of Chandezon et al (the C-method) for modeling gratings reproduces the dip at 23° for our grating structure.

We looked at the reflectivity of p and s polarised light from both structured and polished flat metallic surfaces at this particular angle of incidence (23°), while varying the intensity of incident light over three orders of magnitude. We observe that the grating reflectivity (Fig.2) for s polarisation shows exactly the same behaviour as optically flat Au with the same threshold for plasma formation ($I_{Th}$) in it. The reflectivity of p polarised light from grating shows interesting behaviour. At low intensity it starts from the value 0.33 which, as expected, is roughly the same as the reflectivity of ultrashort pulse assisting SPR. $I_{Th}$ in this case is dramatically reduced by a factor of 3 to a value $1.14 \times 10^{13} W cm^{-2}$ which is quite close to the experimentally observed value. The absorption increases with increasing intensity and at about $2 \times 10^{15} W cm^{-2}$ reflectivity drops by 4.7 times its low intensity value and almost 93% of the incident light is absorbed.

To explain the reflectivity vs laser intensity data in Fig. 2, we use the formalism suggested by Chandezon [5, 6]. The inputs which are needed for this calculation are the grating parameters, which are known, and the refractive index (dielectric constant) as a function of laser intensity. Assuming drude permittivity $\epsilon = 1 - \frac{\omega_p^2}{\omega(\omega + i\nu)}$, we integrate the Helmholtz equation across the density gradient [7]. Here the collision frequency $\nu$ is a function of electron density $n_e$ and electron temperature $T_e$ which in turn depends on laser intensity. Choosing a linear density profile with density scale length ($L/\lambda$) and taking $\nu/\omega \propto n_e$ (following ref. [7]), we calculate the effective collision frequency ($\nu/\omega)_{eff}$ at the solid surface by matching the calculated reflectivity and the measured reflectivity for S-polarized light in plane Au target. From the calculated values of ($\nu/\omega)_{eff}$ at the solid surface as a function of laser intensity, we calculate the dielectric constant (refractive index) at different intensities. To take in to account the effect produced by target (grating) expansion (not included in Chandezon formalism) which actually happens at
high intensities, during the calculation of refractive index, we have modified the imaginary part of dielectric constant \((\nu/\omega)_{\text{eff}}\) by a constant factor \((\sim 0.3)\). The solid lines in Fig. 2 show the results of our calculation. The close match with experiments is a clear vindication of Chandezon formalism.

The absorbed light goes into the plasma and ultimately manifests itself, among other things, as hot electrons which emit bremsstrahlung x-ray and characteristic line radiations. The bremsstrahlung photon energy distribution carries information about the hot electron temperature and the bremsstrahlung yield correspond to the number of hot electrons. The hard x-ray emission spectrum from the plasma generated on the grating surface when it is allowed to interact with intense \((I = 3.8 \times 10^{15} \text{W cm}^{-2})\) s-polarised laser incident at \(\theta_{\text{sp}}\) gives rise to maxwellian hot electron distribution with single temperature component around \(T_h = 16\pm 1\) KeV while for p-polarisation the spectrum is shows non-maxwellian behaviour with two temperature hot electron distribution: one at \(T_{h1} = 14 \pm 3\) KeV and the other at \(T_{h2} = 67 \pm 11\) KeV the higher component constituting 14\% of the total number of hot electrons. The hard x-ray yield for p-polarised light is 4.8 times that of s-polarised case. That, the surface plasmon coupling plays the most crucial role here (in the case of gratings) in the enhanced generation of hotter electrons, is verified when we observe grating-plasma hard x-ray spectrum ( at the same \(I = 3.8 \times 10^{15} \text{W cm}^{-2}\)) at an angle of incidence different from \(\theta_{\text{sp}}\). We don’t observe any hotter component in the bremsstrahlung spectrum when \(\theta_i = 45^0\).

We note that the pit formed (Fig. 1 lower-right inset) due to p-polarisation (at SPR) is bigger and deeper compared to that due to s-polarisation (No SPR) as expected from the reflectivity data. The grating surface surrounding the focal spot is also much more damaged in p irradiated spot, a feature that indicates violent excitation of surface charge density oscillations when the grating is exposed to incoming high intensity ultrashort laser pulse and larger length of propagation of the generated SP wave along the metal vacuum interface.

4. Summary

In summary, we see that it is possible to enhance to a great extent the light coupling in the high intensity regime by exploiting SPR. The reflectivity curves show that the high level of light absorption is maintained even when we increase the light intensity over 3 orders of magnitudes. The lowering of the threshold of plasma formation in grating targets under conditions favourable to SPR is indicative of the increase in the effective field intensity due to the surface plasmon assisted local field enhancement. From the bremsstrahlung radiation at high intensity we observe that SPR is assisting the generation of hotter electrons in larger numbers.

For the present work, the authors GRK and AD acknowledge their respective DAE-SRC-ORI grants from the Government of India.

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