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Role of prepulses in the interaction of intense, ultrashort lasers with "structured" surfaces

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Abstract. We examine enhanced hard x-ray emission (20 – 200 keV) from plasmas produced on nanoparticles coated optically polished copper surface under different prepulse conditions. We observe that enhancement reduces with increasing prepulse intensity. The dynamics of the process is seen to be in the ps regime. We attribute this to preplasma formation on nanoparticles and subsequent modification/destruction of the nanostructure layer before the arrival of the main pulse. It is suggested that high-contrast ultrashort pulses are essential for nanoparticles to function as yield enhancers.

1. INTRODUCTION

In recent years, plasmas generated by intense, ultrashort lasers have attracted multifaceted research to explore basic physics as well as applications. These plasmas are bright and ultrashort sources of x-rays [1, 2]. The radiation pulses are potentially useful in lithography, time-resolved mapping of ultrafast atomic and molecular processes, precision imaging *etc.* [3-5]. Since a practical realization of such sources demands high flux levels, there is a great deal of interest in methods to enhance the x-ray yield and the influence of various laser and target conditions has been the subject of many recent studies [6-9]. Methods of enhancing emission in the very hard x-ray spectral region are being explored only recently. Such studies are also interesting not only from the point of view of the enhanced radiation, but also to understand the role of surface structures or 'roughness' in enhancing the production of hot electrons in plasma responsible for the emission. Enhanced x-ray yield is a signature of enhanced hot electron production, a central issue in inertial fusion research and high energy particle generation and acceleration.

Recently we have proposed that metal nanoparticles (NPs) can enhance the local fields around them, making them excellent sources of hard x-ray pulses [10]. The amount of enhancement depends on the dielectric function of the material and more critically on the shape of the NP, both of which change during the light interaction. The proposed model that explains the phenomena assumes that the inherent local field modification around nanostructures prevail even under intense light fields [10, 11]. Though this is not a serious issue in femtosecond time scales, it might be significant for longer pulses or pulses with prepulses or picosecond pedestal levels. The realization of bright NP X-ray source would involve high intensity lasers, with inherent prepulse and pedestal levels. Thus, it is crucial to learn the effect of prepulse intensity on the emission process.

Here we address this problem in detail by monitoring x-ray emission from optically polished copper surfaces as well as those coated with spherical copper nanoparticles.

2. EXPERIMENTAL

The laser used for our experiments (set up sketched in Fig. 1) is a Ti: Sapphire system (100 fs duration at 10 Hz, 806 nm). The linearly polarized laser beam is split into two and both beams are focused on the

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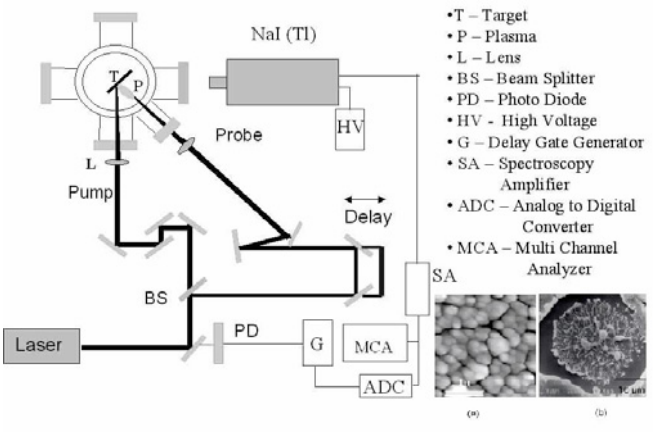

Figure 1. Schematics of the Experimental Set-up. Inset: Scanning Electron Micrographs of (a) the nanoparticle coating and (b) destruction of the nanoparticle layer after exposure to a pulse at the highest intensity used in the study.

targets housed in a vacuum chamber at 10^{-3} Torr, with precise overlap of the focal regions. The target is continuously translated such that each laser shot irradiates a fresh area on the target. The maximum main beam energy is limited to 4.5 mJ in the present series of experiments, yielding a light intensity of about $4.5 \times 10^{15}\,\mathrm{Wcm}^{-2}$ at a focal spot. A variable optical delay is introduced in the weaker beam path using a motorized precision translation stage. We keep the prepulse intensity above the plasma formation threshold ($\sim 10^{13}$-$10^{14}\,\mathrm{Wcm}^{-2}$ for Copper [12]) such that a pre-plasma is formed before the main pulse is incident. To determine the temporal overlap, the prepulse reflectivity (at very low prepulse levels, such that the prepulse does not produce plasma) is monitored as the delay is changed. A sharp drop in the prepulse reflectivity, which is indicative of plasma formation by the main pulse, establishes the "zero" of temporal overlap [13]. X-ray emission from the plasma is monitored using a time-gated NaI (Tl) scintillation detector kept in the plane of incidence at 45^0 to the target normal [14].

Copper NPs are deposited by high-pressure dc-magnetron (Atom Tech 320-O) sputtering [12, 15] on optically polished copper discs maintained at a temperature of 0° C. The nanocrystalline thin films typically consist of a collection of densely packed, spherical NPs, as shown in Figure 1 (a) (inset). The resulting nanocrystalline Cu films are optically flat and 1 μm in thickness. The crystallographic domain size (d_{XRD}) is obtained from x-ray diffraction line broadening. For a film deposited in 180 mTorr Ar environment at a sputtering power of 200W, we obtain $d_{XRD} = 15$nm. Since the thickness of the NP layer is greater than the skin depth at 806nm, the laser essentially interacts only with the film and not the substrate behind it. Figure 1 (b) (inset) is a Scanning Electron Micrograph of one such focal spot. It is evident that the laser destroys just the layer leaving the optically flat copper surface behind it unaffected.

3. RESULTS AND DISCUSSIONS

Figure 2 (inset) presents a typical comparison of bremsstrahlung emission, measured from an optically polished copper surface and such a surface coated with spherical NPs, irradiated at 45^o at $6.0 \times 10^{14}\,\mathrm{Wcm}^2$ with a single pulse. The total energy emitted per pulse from a polished target is 4.2×10^{-14} J while the spherical NPs yield 1.4×10^{-13} J, giving about 3-fold enhancement in the range 20-200 keV. The observed enhanced emission is due to the electric field enhancements near the nanostructures [10, 13]. Figure 2 shows the result of irradiating a spherical NP-coated target with a prepulse of intensity $\sim 10^{14}\,\mathrm{Wcm}^{-2}$ at normal incidence. The intensity of the main pulse was around $10^{16}\,\mathrm{Wcm}^{-2}$. A constant

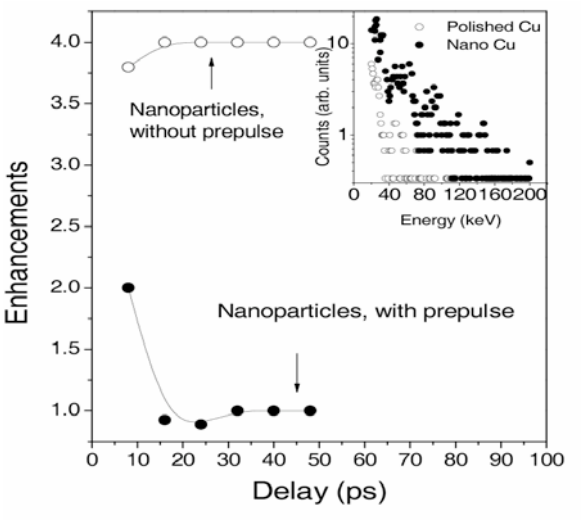

Figure 2. Effect of a prepulse on nanoparticle-coated targets at various delays. The enhancement factor (ratio of the x-ray yield of nanoparticle coated targets to that from uncoated targets) reduces drastically and vanishes with the introduction of a prepulse at $\sim 10^{14}\,\mathrm{Wcm}^{-2}$. Inset: Typical hard x-ray spectrum from a polished copper target and a nanoparticle-coated target.

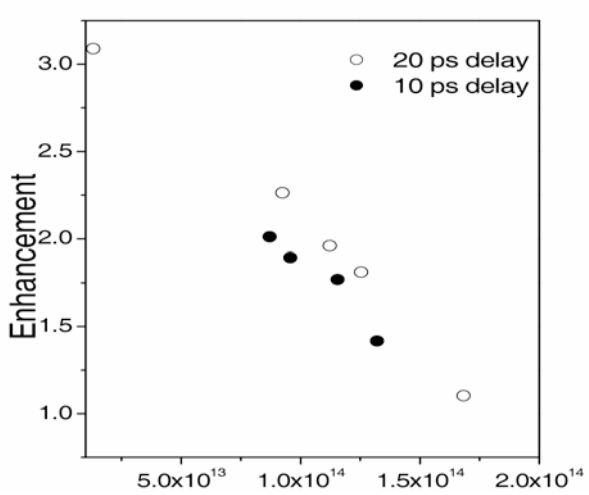

Figure 3. Variation of the x-ray yield enhancement with prepulse intensity and delays. The main pulse intensity is $\sim 5 \times 10^{15}\,\mathrm{Wcm}^{-2}$.

4-fold enhancement was obtained from the NP-coated surface, when irradiated with just the main pulse, without the prepulse, similar to the result shown in the inset. This is obviously not a function of the prepulse delay but the plotted data points (top curve) indicate just the values obtained with different measurements. As is clear from the figure, there is a drastic drop in the enhancement on irradiation of a prepulse, even at small delays ($\sim$10ps). The enhancement vanishes completely as the delay between the prepulse and the main pulse increases.In the case of nanostructured surfaces, preplasma formation affects the interaction detrimentally, as discussed above.

It is important to parameterise conditions at which nanostructures provide enhancements in x-ray yield. We have studied the x-ray production from nanoparticle-coated targets and flat copper targets under various prepulse levels and delays between the prepulse and the main pulse (Figure 2). Yield enhancements from nanoparticles at main pulse intensities $\sim 4.5 \times 10^{15}\,\mathrm{Wcm}^{-2}$ and under different prepulse levels show that the enhancement starts reducing monotonically once the prepulse levels exceed $10^{13}\,\mathrm{Wcm}^{-2}$. It takes a prepulse intensity $\sim 2 \times 10^{14}\,\mathrm{Wcm}^{-2}$ to completely nullify the effect of nanoparticles. Lower intensities do not seem to alter or destroy the nanostructures to an extent that their effect is removed completely in the interaction with the following ultrashort pulse.

4. SUMMARY

In summary, we show that prepulses can significantly reduce or even nullify the enhanced absorption offered by the nanostructures. In ultrashort pulse laser-solid interaction the nullification at appropriate prepulse intensities ($10^{14}\,\mathrm{Wcm}^{-2}$) is observed to occur on picosecond timescales. These observations are especially important in studies to find efficient laser plasma sources of short wavelength radiation. The prepulses and pedestals invariably associated with high power lasers can be detrimental to the enhanced absorption induced by the nanostructures. High contrast pulses are essential in this respect.

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