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Nanosecond and femtosecond pulsed laser ignition of reactive multilayer foils

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ABSTRACT

Pulsed laser ignition thresholds for Al/Pt energetic multilayer foils are characterized as a function of multilayer design. A comparison of ignition thresholds for 30 nanosecond (ns) and 100 femtosecond (fs) pulses is presented, the results of which demonstrate differences between ns and fs pulsed laser-material interactions. Specifically, the ns pulsed laser ignition thresholds are less than the fs ignition thresholds when comparing multilayers of identical periodicity and total thickness. We attribute the differences in ignition behavior to the thermal character of ns-pulse interactions with a solid versus the limited thermal diffusion and subsequent ablation of laser heated material associated with fs pulse interaction.

INTRODUCTION

Energetic multilayer foils are typically grown via sputter deposition and consist of distinct layers of two materials which react exothermically [1]. If sufficient heat is generated via local stimulation (ignition), surrounding material can be heated and caused to mix, generating a runaway reaction which can propagate throughout the entire foil. Examples of exothermic material systems that exhibit self-propagating high temperature synthesis (SHS) reactions include Ni/Al, Co/Al, Ni/Ti, and Al/Pt. These materials have found use for specialized joining applications [2,3], and for the controlled synthesis of exotic compounds via SHS [4].

While electronic matches or a simple spark from a 9 Volt battery can be used to ignite these foils, laser ignition provides a unique opportunity to characterize ignition processes with a very clean and reproducible energy source. Additionally, laser cutting has been proposed as a method for fabricating odd-shaped foils for joining [5]. In a previous work, the nanosecond (ns, 10^{-9}) pulsed laser ignition threshold was studied for three material systems, specifically Co/Al, Ni/Ti, and Al/Pt, with the goal of identifying the influence of material properties on ignition [6]. In the present work, the influence of laser pulse duration on ignition behavior is investigated for Al/Pt multilayer reactive foils. While providing important information on ignition behavior of reactive foils, interesting insights into the laser-material interaction for ns and femtosecond (fs, 10^{-15}) pulsed irradiation are also obtained.

Detailed studies of fs and ns pulsed laser – material interactions have been made with a wide range of in-situ and ex-situ methods [7-14]. From these previous studies, it has been observed that the mechanisms for material removal (or ablation) changes as a function of laser pulse duration[7,9,10,12,14]. The evolving mechanisms for ablation results in a reduction of heat-

affected zones (HAZ) and increased reproducibility for fs laser ablation relative to that produced by ns pulsed lasers [7,10,15,16]. The ignition threshold relationship for 100 fs and 30 ns laser pulses observed here is attributed to the differences in the laser material interaction as a function of pulse duration.

EXPERIMENT

All multilayer films are initially grown on Si(100) substrates with 400 nm thermal oxide via direct current (DC) magnetron sputtering. The base pressure of the growth chamber is 10^{-8} Torr, and sputtering is performed with a process gas of Ar at pressures of 10 mTorr. Two general characteristics define the physical features of a film: the total thickness of the film, and the bilayer thickness of the constituent layers. The total thickness of the Al/Pt films is 1.56 μm for all films studied. The bilayer thickness is the sum of the thickness of an individual layer of each representative material in the film. For example, a particular bilayer thickness of Al/Pt is 195 nm (Al = 102 nm, Pt = 93 nm), and 8 total bilayers are stacked in order to create a total film thickness of 1.56 μm. While the total film thickness is held constant, the bilayer thickness is varied over a range from 10 – 200 nm.

Following deposition, foils are carefully removed from their substrates, and cut into approximately 5 mm x 1 mm strips that are then suspended across the inner diameter of a washer for laser ignition tests. During this procedure, care was taken to consistently place the Pt surface upward such that laser pulses encountered the same material and therefore a constant optical reflectivity could be estimated. An optical intensity reflectivity coefficient of 0.716 for Pt at a wavelength of 800 nm (1.5 eV photon energy) [17] was used to correct the ignition threshold values.

The laser used for ignition threshold measurements is a Ti:sapphire laser with a temporal pulse width of 100 fs centered at a wavelength of 800 nm. To obtain 30 ns pulses from the laser, the injection of the fs seed pulse is blocked before it enters the regenerative amplifier, such that lasing is achieved on the pump pulse alone. A 10 cm focal length lens is used to focus laser beam onto the foil surface, yielding a focused beam diameter of 27.2 ± 0.4 μm for both pulse durations with a Gaussian spatial intensity profile. The laser is operated in a single shot mode, with ignition determined for a particular foil by incrementally increasing the laser pulse energy (via a quarter waveplate and polarizer) until an audible “pop” or visible flash indicates that ignition has occurred. Note that single shot experiments were spaced across each foil to avoid cumulative effects of damage, melting, etc. The ignition threshold from at least 10 foils pieces were measured for a particular bilayer thickness and pulse duration. All pulsed laser ignition threshold measurements were made in air at ambient conditions. The ablation threshold of a 200 nm Pt film on a Si(100) substrate was also measured for both pulse durations using standard procedures [18].

DISCUSSION

The ignition thresholds for Al/Pt multilayer foils as a function of bilayer thickness for both 100 fs and 30 ns pulse durations are shown in Figure 1. Also plotted in Figure 1 is the net reaction velocity obtained from high speed photography conducted far from the ignition zone [6]. Three observations are made from the data presented in Figure 1. First, the ignition threshold for both pulse durations varies with bilayer thickness, gradually increasing with increasing bilayer

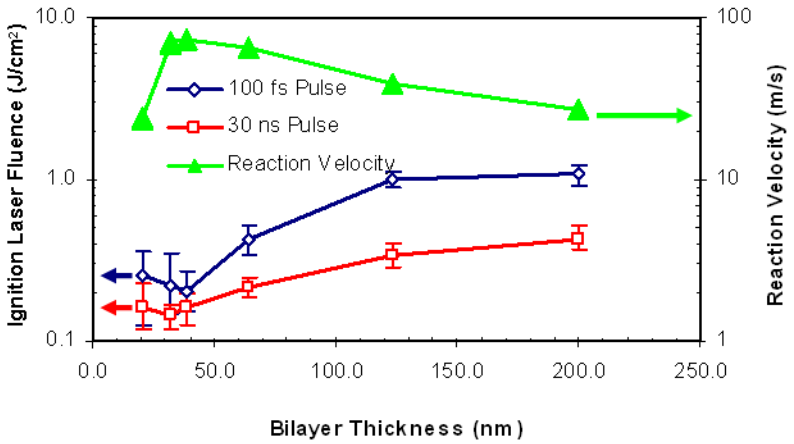

Figure 1: Pulsed laser ignition thresholds and net reaction velocity for Al/Pt multilayer reactive foils as a function of bilayer thickness. Ignition results have been corrected for the linear reflectivity of a Pt surface at a wavelength of 800 nm (R = 0.716).

thickness. In general, the reaction of exothermic materials is limited at large bilayer thicknesses by mass diffusion, i.e. the length to which atoms of each chemical species must travel in order to combine and release heat [19]. As the bilayer thickness is increased, material from neighboring layers must travel further and further to mix which effectively decreases the velocity of the reaction. This presumably is also the reason why the ignition threshold increases with increasing bilayer thickness for bilayer thicknesses exceeding approximately 40 nm. Second, for bilayer thicknesses less than ~ 40 nm the velocity decreases; this has been attributed to the effects of premixed reactants in as-deposited foils [4,20]. The ignition laser fluences (ns and fs) do not continue to decrease in this regime with diminishing bilayer thickness. The third observation to be drawn from Figure 1 is that the ignition threshold for 100 fs generally exceeds that for 30 ns when bilayer thickness is fixed.

Two possible explanations are proposed to explain the ignition threshold relationship between 100 fs and 30 ns laser pulses (our third observation). First the HAZ associated with the ns pulsed laser-material interaction provides sufficient mixing of material to ignite a reaction. This is in contrast to the HAZ associated with fs pulsed laser-irradiation which is quite limited [7,10,15,16] and does not provide sufficient mixing to ignite a reaction until much higher fluences are reached. As evidence for the presence or absence of HAZ, consider the optical microscope images of Figure 2 where single shot ablation features are shown for both 30 ns and 100 fs laser pulses on a 200 nm Pt film. The ablation features produced with the 30 ns pulses

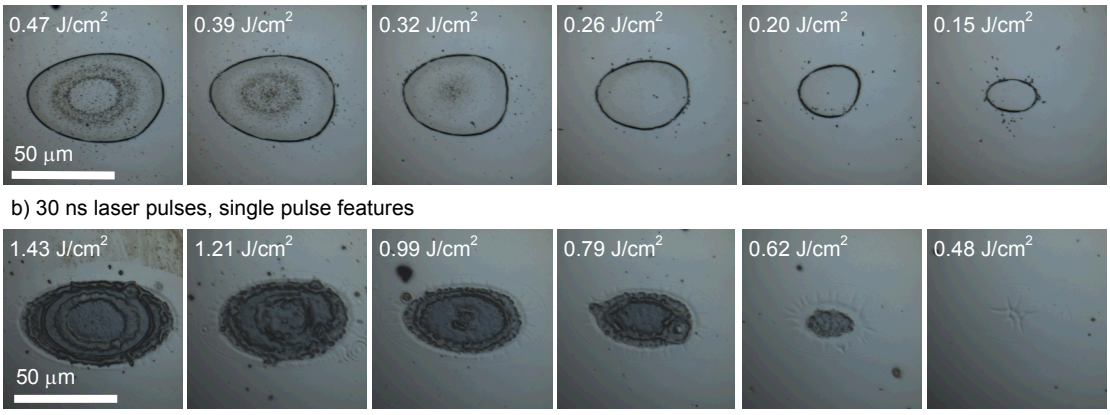

Figure 2: Single pulse ablation features produced with a) 100 fs and b) 30 ns laser pulses at a wavelength of 800 nm with indicated laser fluence. The scale bar in the left most images of each series apply to all images in the series.

exhibit significant recast molten material, apparent due to the darker, material at the center of the features down to the lowest laser fluences for which ablation is observed. This in the contrast to the 100 fs ablation features where no such molten material is present, and the ablation crater is smooth and well defined down to the lowest laser fluences for which ablation is observed. Similar observations have been widely reported for ablation of variety of metals with fs and ns laser pulses [7,10].

The effect of material ablation is the second proposed phenomena underlying the increased ignition threshold in the case of fs pulses. Comparing Figures 1 and 2, one notes that no detectable ablation occurs during ns-laser ignition (at threshold), yet fs-laser ignition is accompanied by some ablation for all designs tested. The measured ablation threshold for a 200 nm Pt film for 30 ns laser pulses is 0.49 ± 0.026 J/cm^2, while the ablation threshold for 100 fs laser pulses is 0.14 ± 0.003 J/cm^2. While the ablation threshold of the 30 ns pulse is expected to be greater than that for 100 fs laser pulses [10,14], the fact that only fs-laser ignition is affected by ablation is recognized as being potentially important. For ablation with fs laser pulses, particularly near the threshold for ablation, the material initially energized by the laser energy undergoes a rapid transition from room temperature to a superheated mixture of plasma and liquid droplets [21,22]. The shock associated with rapid thermal expansion of material [23,24] reflects off the underlying solid material resulting in the ejection of the overlying superheated layer [21,25]. This mechanism occurs on a timescale (~ 1 ns) that limits thermal transport from the laser-heated material into the surrounding, relatively cool material [21,26,27]. Furthermore, very little ballistic transport of energetic carriers from the laser-heated volume to the surrounding relative cool bulk material, which further limits the potential for heating surrounding material [28]. These processes combined lead to the sudden ejection or ablation of all laser-heated material and limit thermal diffusion into the surrounding volume, thereby suppressing ignition that relies on heat to mix the constituent materials and generate a runaway exothermic reaction.

It should be noted that at higher laser fluences (typically around 0.4-5 J/cm^2 depending on the material) ablation by fs laser pulses becomes more thermal in character as ballistic electrons excited by the laser irradiation have sufficient energy and density to propagate into material outside of that initially excited by the laser [28-30]. This leads to an ablation mechanism more similar to boiling and evaporation of material observed with ns laser ablation [31,32]. As a result, we speculate that exothermic material systems which exhibit a higher general ignition threshold (such as Co/Al or Ni/Ti [6]) may have ignition thresholds which are less dependent on the pulse duration of the laser source.

CONCLUSIONS

The single-pulse ignition thresholds of energetic multilayer foils comprised of Al and Pt were studied as a function of bilayer thickness for 100 fs and 30 ns laser pulse durations. The ignition thresholds for both pulse durations are found to vary with bilayer thickness, in a trend inversely related to the measured reaction velocity, suggesting that effects of mass transport which limit reaction propagation also increase the energy required to produce ignition. When testing a foil having a given bilayer thickness (multilayer periodicity), the ignition threshold is observed to be greater for 100 fs laser pulses when compared with longer 30 ns laser pulses. This difference is not yet fully understood, but may be related to greater thermal effects associated with ns pulsed laser-material interaction, or to ablation of hot material before thermal transport can occur under the case of fs pulsed laser-material interaction.

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