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Pulsed Laser Ignition of Reactive Al/Pt Multilayer Foils

Joel P. McDonald and David P. Adams

Sandia National Laboratories, Albuquerque, New Mexico 87185, USA

Abstract. Pulsed lasers were used to study the ignition behavior of reactive multilayer foils comprised of Al and Pt. Specifically, ignition threshold were characterized as a function of foil properties, laser pulse duration, and focused laser beam diameter. The ignition threshold as a function of foil bilayer thickness exhibits trends consistent with mass diffusion limited reactions present in energetic/reactive multilayers. Furthermore, it is proposed that evolving physical and thermal characteristics of laser-material interactions as a function of laser pulse duration have a direct influence on ignition phenomena.

Keywords: femtosecond, nanosecond, laser ignition, reactive foils
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INTRODUCTION

Reactive multilayer films or foils are typically comprised of two or more materials that react exothermically when they are caused to mix [1]. The heat released by the exothermic reaction can be used for applications including joining [2] and for the preparation of novel material phases [3, 4]. Fundamental to the utility of reactive multilayers is the manner in which the exothermic chemical energy is stored for future use. The reactive constituent materials are deposited at low temperature in distinct layers, such that under normal environmental conditions the constituents do not mix and therefore do not release energy from the exothermic chemical reaction. When the reactive multilayer foil is ready for use, an impulse of some form is used to initiate local mixing. Should this impulse provide sufficient mixing of the constituents, the associated exothermic reaction can initiate mixing of surrounding constituent layers, propagating the reaction throughout the foil [1]. A wide variety of impulses can be used to ignite reactive multilayer foils, including electrostatic discharge [5], and the focus of this investigation, pulsed laser irradiation [6, 7].

Previous works have investigated the nanosecond (ns) pulsed laser ignition of Al/Pt, Ni/Ti and Co/Al reactive multilayer foils [6, 7]. These works primarily focused on the role of reactive foil parameters on the laser ignition. For example, it was found systems possessing greater stored energy were easier to ignite, with the pulsed laser ignition threshold decreasing with increasing magnitude of the enthalpy of formation (note that the enthalpy of formation is always negative for exothermic reactions). Furthermore, the ignition threshold was found to vary with the thickness of the layers of the constituent materials (called the *bilayer thickness*), although no discernable trend could be indentified.

Here, ignition of reactive multilayers is investigated for Al/Pt foils as a function of both foil and laser parameters. First we report the ignition threshold as a function of bilayer thickness (the thickness of an individual pair of Al and Pt layers) for 90 femtosecond (fs) and 30 ns laser pulses. Second, the ignition threshold as a function of

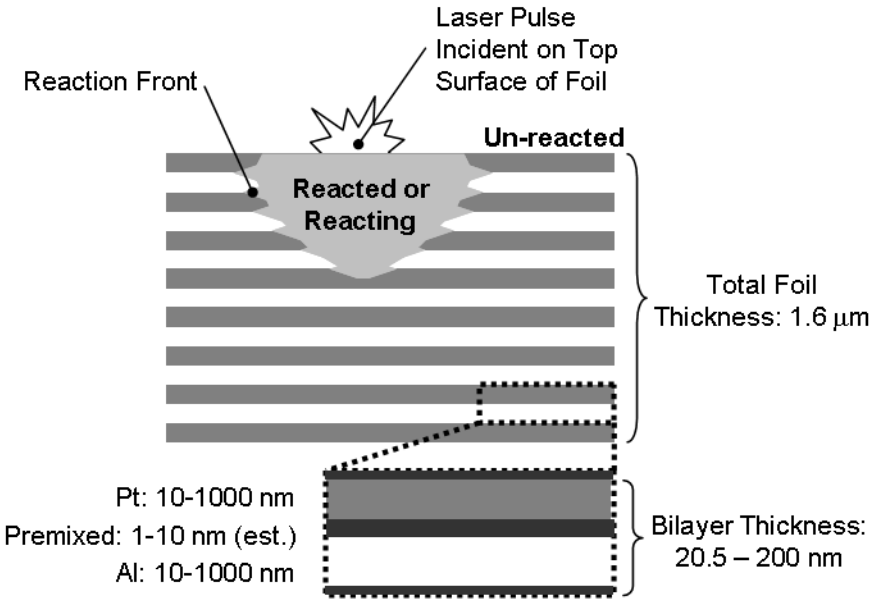

FIGURE 1. Schematic cross section of an Al/Pt multilayer reactive foil. For pulsed laser ignition experiments, the laser was always incident on the Pt surface of the multilayer.

focused laser beam diameter is presented for a particular Al/Pt foil for both 100 fs and 30 ns laser pulses. Finally, the ignition threshold of a particular Al/Pt foil is presented over a broad range of laser pulse durations (100 fs – 50 ms), indicating a sharp transition in ignition phenomena as the pulse duration enters the microsecond (s) domain. The results of these investigations bring to light important aspects of ignition phenomena in reactive multilayer foils while providing details of laser-material interactions consistent with previous studies as a function of laser pulse duration.

EXPERIMENT

All multilayer foils were prepared with direct current (DC) magnetron sputtering. Here, foils comprised of alternating Al and Pt layers were studied. The bilayer thickness, or thickness of a single pair of Al and Pt layers, was varied from 20.5 – 200 nm. As grown, the total thickness for all films was 1.6 m. For laser ignition experiments the films were removed from their oxidized Si(100) substrates and ~ 1 mm by 5 mm sections of each multilayer foil were prepared. Note that once multilayer films have been removed from their substrates, they are referred to as multilayer *foils*. A schematic of a reactive foil cross section is shown in Figure 1.

Two lasers were used for ignition threshold measurements. A chirped pulse amplified Ti:sapphire laser (Spectra Physics, Hurricane) with a central wavelength of 800 nm was used to produce laser pulse durations (FWHM) of 90-100 femtoseconds (fs), 1.0 ± 0.08 picosecond (ps), and 30.0 ± 2.0 nanoseconds (ns). The native pulse duration coming from the Ti:sapphire laser was ~100 fs (measured via autocorrelation), and to obtain an ~1 ps pulse (measured via autocorrelation) the re-compression of the amplified pulse was degraded by changing the grating position within the regenerative amplifier. To obtain a ~30 ns pulse from the laser (measured

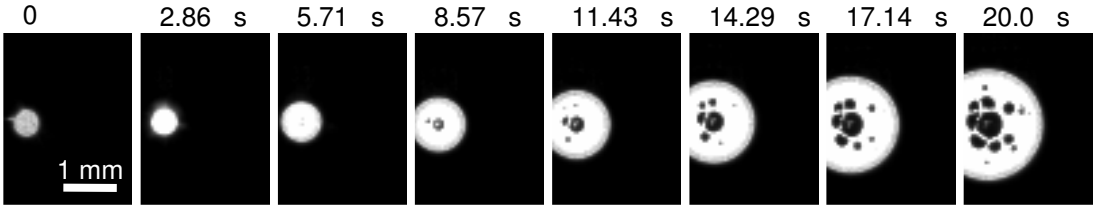

FIGURE 2. High speed photography of a reaction in an Al/Pt multilayer reactive foil with a bilayer thickness of 123.1 nm following single pulse laser ignition of an at a laser fluence 0.96 ± 0.05 J/cm^2.

via photodiode and oscilloscope), the fs seed pulse was blocked from entering the regenerative amplifier and lasing was achieved from the pump pulse alone. High speed images of ignition and subsequent reaction of an Al/Pt multilayer with single 100 fs laser pulse is shown in Figure 2.

A fiber coupled diode laser (Northrup Grumman) at a central wavelength of 808 nm was the second laser used for ignition threshold measurements. The pulse duration was electronically controlled over the range from 70 microseconds (s) to 50 milliseconds (ms) measured via photodiode. As indicated in Figure 1, the Pt surface of the foil was exposed to the laser for all ignition threshold measurements in order to maintain consistent surface reflectivity between measurements. Note that all stated values of laser fluence within this work have been reduced by the linear intensity reflectivity of the Pt surface (0.72 [8]). For each data point, at least 7 independent ignition threshold measurements were made. The error bars shown in plots represent the maximum and minimum values obtained for the ignition threshold of each foil.

RESULTS/DISCUSSION

Ignition threshold measurements as a function bilayer thickness for two laser pulse durations (100 fs and 30 ns) are shown in Figure 3. For these measurements, the laser was focused to a beam radius (Gaussian, 1/e^2) of 13.6 ± 1.0 m on the foil surface. It is observed that the ignition threshold generally increases for both pulse durations once the bilayer thickness exceeds 39 nm. It is also apparent that the average reaction propagation speed decreases over this same range of bilayer thicknesses. It is expected that these trends result from the same general phenomenon; as the bilayer thickness increases, the atomic diffusion length required to propagate the exothermic reaction also increases. As a result, the onset and subsequent propagation of the reaction is inhibited [7]. The reaction speed decreases for very small bilayer thicknesses as well (for example 20.5 nm), as the naturally occurring pre-mixed AlPt present at the interface between layers (shown in Figure 1) becomes a significant fraction of the total foil volume. As the pre-mixed material is already in some compound phase, it is no longer a significant source of exothermic energy, thereby limiting the energy available to propagate a reaction [9]. It is proposed that the apparent leveling off of the ignition threshold for small bilayer thickness (< 39 nm) is due to the same reaction-limiting effects of the pre-mixed AlPt.

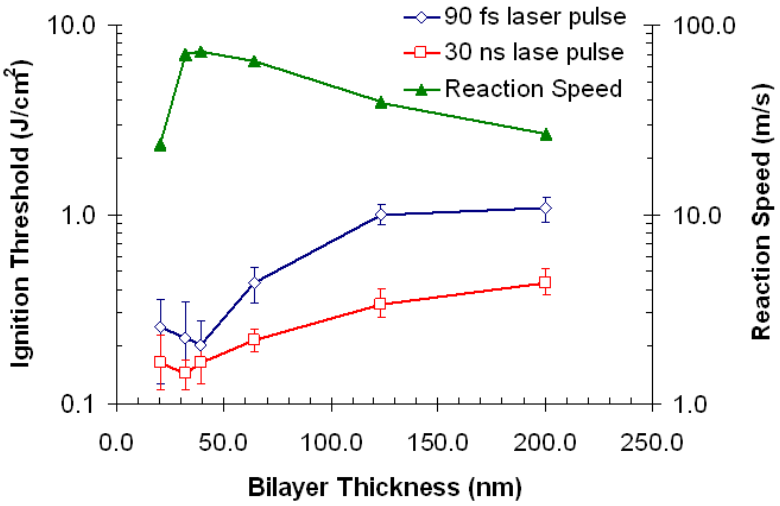

FIGURE 3. Single pulse laser Ignition threshold and reaction speeds of Al/Pt multilayer reactive foil as a function of bilayer thickness. The Pt surface was exposed to the laser pulse for all ignition threshold measurements. The error bars represent the maximum and minimum threshold values obtained for each point.

For the particular focused laser beam radius of 13.6 ± 1.0 m, the average ignition threshold for 90 fs laser pulses is greater than that for 30 ns laser pulses for all bilayer thicknesses studied. This relationship is attributed to the emerging role of thermal processes associated with increasing pulse duration in this regime [10]. In general, material heating by the action of the laser pulse is presumed the dominant mechanism underlying laser ignition and subsequent reaction propagation in reactive multilayer foils. For irradiation with ultrashort pulses (< 1-5 ps [11]), metallic materials proceed through a rapid transition from the solid to the vapor phase, producing an expanding plasma which carries the ablating material away from the surface before heat can be transported out of the initially energized volume into surrounding, non-ablating material [10, 11]. As a result, very little transient heating is produced particularly at laser fluences near the threshold for ablation, which effectively inhibits ignition by the action of the laser pulse. The laser-material interaction associated with ns duration laser pulses allows sufficient time for thermal diffusion into regions outside the volume of material initially excited by the laser pulse[10]. The transient heating and heat affected zone facilitate ignition of reactive multilayer foils with ns duration laser pulses.

Qualitative evidence for the different thermal characteristics of damage produced by fs and ns laser pulses is presented in Figure 4, where single shot ablation features as a function of laser fluence are shown for both 90 fs and 30 ns laser pulses. These images reveal that significant *recast* molten material with high reflectivity is present for 30 ns pulse ablation for all laser fluences, while similar recast (splattered) molten material is only observed at higher laser fluences (~ 5 times the ablation threshold) for 90 fs laser pulses [11]. To measure the ablation threshold of the 5 m Pt film, a series of similar images were acquired as a function of laser pulse energy, yielding values of 0.11 ± 0.004 J/cm^2 and 0.44 ± 0.03 J/cm^2 for 90 fs and 30 ns pulses respectively (values corrected for the reflectivity of the Pt surface [8]). As such, for 90 fs laser

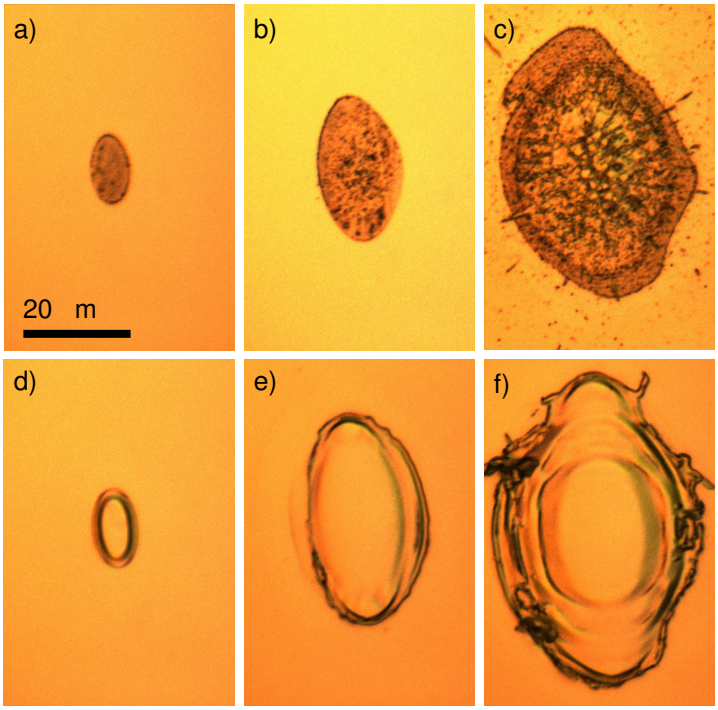

FIGURE 4. Single pulse laser damage features produced on a representative 5 micrometer thick Pt film on Si(100) for pulses of duration and laser fluence: a) 90 fs, 0.2 ± 0.002 J/cm^2, b) 90 fs, 1.0 ± 0.01 J/cm^2, c) 90 fs, 18.4 ± 0.2 J/cm^2, d) 30 ns, 0.58 ± 0.01 J/cm^2, e) 30 ns, 2.6 ± 0.05 J/cm^2, f) 30 ns, 13.0 ± 0.3 J/cm^2. The scale bar in a) applies to all images.

pulses the ignition threshold is equal to or exceeds the ablation threshold for all bilayer thicknesses. On the other hand, for 30 ns laser pulses the ignition threshold is less than or equal to ablation threshold for all bilayer thicknesses. These relationships as a function of laser pulse duration support the view that the ignition threshold of the Al/Pt multilayer is enhanced by the thermal characteristics of the laser-material interaction associated with ns duration laser pulses.

The pulsed laser ignition threshold as a function of the focused beam diameter is presented in Figure 5. For this experiment the focused beam radius (1/e^2) was varied by using lenses with different focal lengths. The spot size was subsequently determined for each lens from ablation threshold measurements[12] performed on Si(100) with 500 nm of PETEOS silicon dioxide. For 30 ns laser pulses it is apparent that the ignition threshold is approximately the same for all focused beam radii, but exhibits increasing variability with decreasing beam radius. For 100 fs laser pulses, the data suggests an increasing ignition threshold as the focused laser spot radius is decreased. To explain the increase in the pulsed laser ignition threshold with decreasing beam radius, one might consider the volume of energized material and associated magnitude of exothermic energy release required for ignition of a reactive multilayer foil. It is expected that the depth of the laser-material interaction will need to increase as the lateral dimensions are decreased in order to preserve a critical amount of energy release and subsequent ignition [7]. This may explain the increase in the laser fluence required for ignition as the spot size is decreased for the data collected with the 100 fs laser pulse duration. Just why the 30 ns laser pulse duration does not exhibit a similar trend is not yet understood, and further investigations into

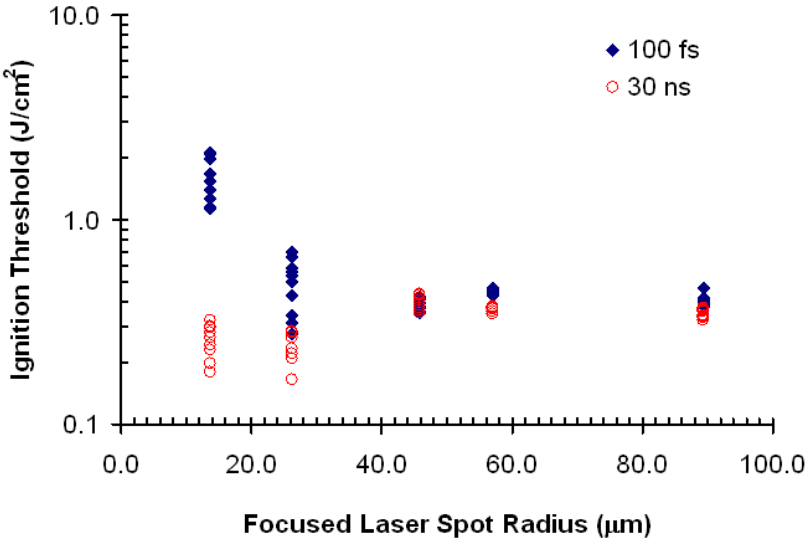

FIGURE 5. Single pulse laser Ignition threshold of an Al/Pt multilayer reactive foil with 123.1 nm bilayer thickness as a function of the focused laser spot radius. The focused laser spot radius for the incident Gaussian beam was determined from ablation threshold measurements. The Pt surface was exposed to the laser pulse for all ignition threshold measurements.

the role of focused beam radius on ignition behavior are underway to address this discrepancy. As a demonstration of the consistency of ignition phenomena, note that the relative ignition threshold values at the ~13.6 m beam radius are very similar (± 10 %) to those presented in Figure 3, in spite of the fact that these data were collected in a different optical setup, on a different day, and with a sample from a different production cycle (but with the same bilayer thickness of 123.1 nm).

The ignition threshold of a specific Al/Pt reactive foil with a bilayer thickness of 123 nm as a function of laser pulse duration from 100 fs to 50 ms is shown in Figure 6. For these measurements, two different lasers were used however the focusing conditions were made consistent, yielding a focused laser spot size of 190 ± 10 m on the foil surface. For pulse durations greater than 100 s the ignition threshold increases uniformly. In general, pulsed laser ignition of reactive multilayers is expected to be dependent on the intensity of the laser pulse, such that the energy in the pulse (and correspondingly the laser fluence) must increase with the laser pulse duration in order to reach the ignition threshold. This is because the so called *ignition temperature* must be reached in some volume of material in order for ignition to occur [1]. Due to thermal diffusion, if the laser pulse energy is delivered too slowly the heat imparted to the system will spread out of the laser energized region such that the ignition temperature is never reached [13]. As a result, as the laser pulse duration is increased, the laser fluence must correspondingly increase by some factor in order for the ignition temperature to be achieved in the required volume.

The apparent leveling off of the ignition threshold for pulse durations less than 100 s is not yet fully understood. For ultrashort pulses (< 10 ps), the onset of ablation may have the moderating effect of increasing the ignition threshold in spite of increases in intensity with decreasing pulse duration. In the case of ablation, the material energized by the laser is removed from the system so that heat propagation

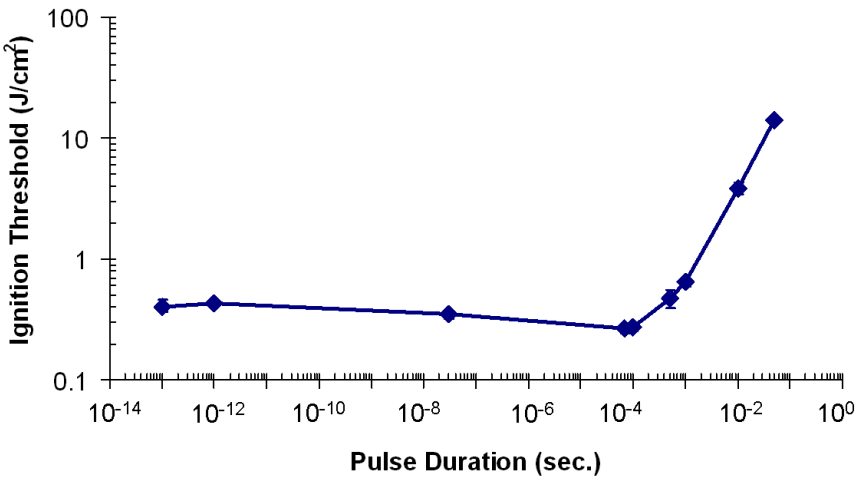

FIGURE 6. Single pulse laser ignition threshold of an Al/Pt multilayer reactive foil with 123.1 nm bilayer thickness as a function of laser pulse duration. The Pt surface was exposed to the laser pulse for all ignition threshold measurements. The error bars represent the maximum and minimum threshold values obtained for each point.

into surrounding material does not occur, effectively inhibiting ignition. The precise mechanism underlying the apparent leveling off of the pulsed laser ignition threshold at pulse durations near 100 s will be the topic of future modeling efforts.

CONCLUSION

Pulsed laser ignition thresholds for Al/Pt reactive multilayer foils were characterized as a function of multilayer design and laser conditions. As a function of foil bilayer thickness, the measured ignition threshold exhibited trends consistent with mass diffusion limited reactions for coarse bilayers and pre-mixed material limited reactions for fine bilayers. It was also observed that for a modest focused beam diameter of 13.6 ± 1.0 m, the ignition threshold for 90 fs laser pulses exceeded that for 30 ns laser pulses for all bilayer thicknesses. Furthermore, the ignition threshold for 90 fs laser pulses exceeded the ablation threshold for all multilayer foils. This particular feature enables joining applications for reactive multilayer foils by demonstrating that a fs pulsed laser can be used to cut and prepare complex shapes in these energetic materials without igniting them.

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