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Nanosecond laser-induced shock propagation in and above organic liquid and solid targets

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ABSTRACT

The study of shock propagation in air and liquid can play an important role in understanding light-matter interactions during laser processing experiments. In this work, we perform plume shadowgraphy experiments on liquid and solid targets of acetone and toluene and calculate the velocity and pressure at the leading edge of the shock front. Our results are compared to recent work in which early blast wave dynamics are studied and the applicability of the classical Taylor–Sedov model is assessed for our data. We observe an enhanced vertical expansion in the shockwave that is attributable to absorption and heating above the surface.

1. Introduction

Toluene and acetone are examples of organic solvents that are frequently used in laser processing experiments such as matrix-assisted pulsed laser evaporation (MAPLE) [1–3], and laser ablation in liquids (LAL) [4–7] because of their versatility. Both can solubilize a wide range of polymers and other materials, and have relatively low reactivity and health concerns. Toluene has been frequently used as a MAPLE matrix [8,9,3,10], whereas acetone is a common choice for both MAPLE [11] and LAL experiments [12–14]. In this work, we utilize the fourth harmonic of a Nd:YAG laser (266 nm) in order to ablate liquid and solid surfaces of acetone and toluene under ambient conditions. The plume is imaged by shadowgraphy and the position of the shockwave is recorded as a function of time. From these measurements, we can determine the velocity and pressure at the leading edge of the shock front.

The Nd:YAG laser used to initiate the shockwave (Ekspla NL-303, 4 ns) was operated in single-shot mode with a pulse energy of 2.25 ± 0.25 mJ and a beam radius of 500 μm. This translates into a fluence of 290 mJ/cm^2, which is typical for UV-MAPLE experiments [15–18,2]. The beam profile was found to be nearly TEM_{00} and the 266 nm radiation was brought to a focus with a 150 mm focal length plano-convex lens. A second 532 nm Nd:YVO_4 laser (Ekspla NL-201) was incident upon a cuvette of Rhodamine dye that illuminates the target surface at a fixed time after the 266 nm pulse arrives, thereby serving as the camera's flash. The time between laser pulses was controlled by a four-channel digital delay generator (Quantum Composers), and the total optical jitter appears to be about 20 ns, which we take to be the temporal resolution of our system. A Pike F-145J or Sentech STC-TB202USH-ASH CCD camera was utilized with a macro lens to record the shadowgraphs. HPLC grade toluene and acetone (Sigma–Aldrich) was obtained and used without further purification. A schematic of the experimental apparatus is shown in Figure 1.

Images were analyzed using the measuring tool in GIMP (Gnu Image Manipulation Program) or ImageJ, and a scale was established either as 2.21 μm/pixel or 6.84 μm/pixel, depending on the camera system. The shock wave expansion was determined in both the horizontal and vertical directions with numerical analysis performed using Microcal Origin.

2. Results and discussion

2.1. Liquid targets: expansion in air

Plume shadowgraphs were recorded for liquid and solid targets of acetone and toluene. Images were separately captured for shockwaves traveling under the liquid surface. In all cases, a single frame was captured for each image. Representative images for shockwaves in air above acetone are shown in Figure 2a–d. These images have much in common with previous reports of ns ablation of liquids [19–21] and organic matrices [22]. There is a well-defined shock wave and a clearly visible Mach stem at the base, see Figure 2b. The shockwave eventually propagates out of frame

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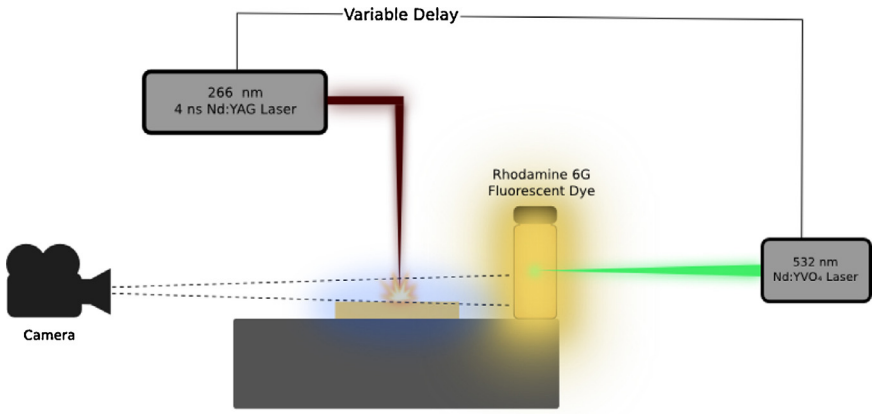

Figure 1. Schematic of shockwave imaging apparatus.

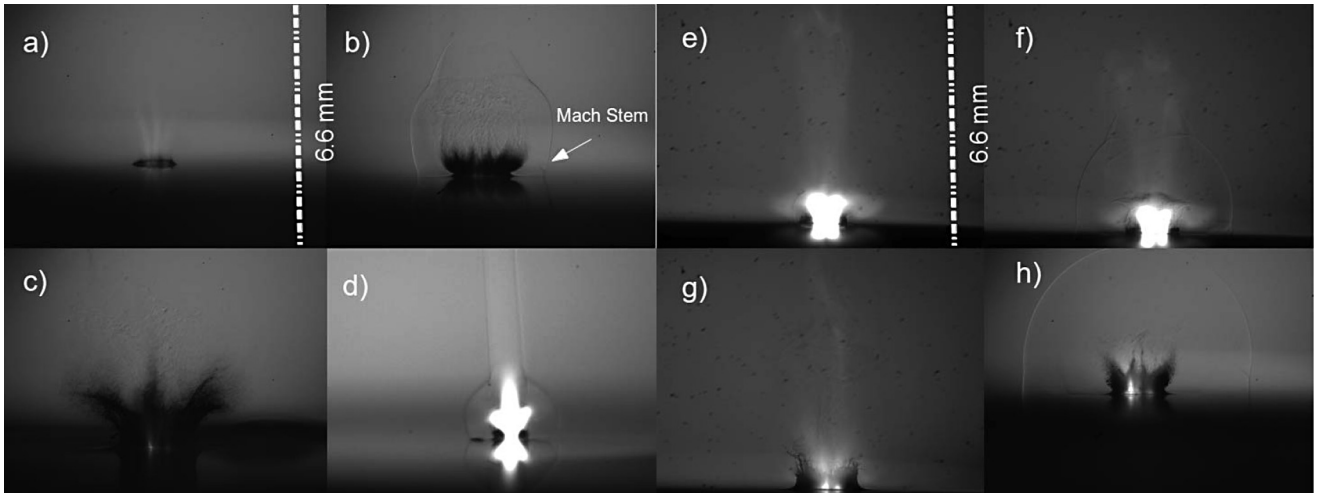

Figure 2. Plume shadowgraphs above liquid acetone for (a) 0 ns delay, (b) 500 ns, and (c) 5000 ns. In (d) we display an image for 500 ns delay with a smaller beam diameter (~280 μm) and lower pulse energy (~2.0 mJ). The vertical column in the image indicates the path of the laser beam. Images above liquid toluene are shown in (e) 0 ns delay, (f) 500 ns, and (g) 5000 ns. In (h), the plume for a non-absorbing liquid (THF) is shown for a delay of 1 μs.

and the toroidal vapor cloud is eventually overtaken by strong recoil-induced ejection that closely resembles droplet dynamics at later times.

One significant difference observed in our results is the presence of a vertical column, see Figure 2. The feature is present in most of the image but it is clearest in frame (d) where the spot size has been reduced and the tilt angle of the camera adjusted. We attribute this feature to strong absorption of the laser by vapor rising from the volatile target. This produces a 'chimney' effect which results in a faster vertical than horizontal propagation of the ablation shock wave that will be discussed further in a later section. A similar result was observed in ablation of toluene with 248 nm laser pulses [23].

Toluene and acetone both absorb 266 nm radiation with moderate strength. In acetone, the absorption band is the result of a $n-\pi^*$ excitation, while for toluene it results from the stronger $\pi-\pi^*$ transition. In both cases, 266 nm is close to the band maximum. Important optical properties for toluene and acetone are listed in Table 1. For comparison, an image was recorded for tetrahydrofuran (THF), a non-absorbing liquid at λ = 266 nm, which does not display the chimney effect.

In Figure 3, we display the early stages of the acetone plume. Initially, the shockwave is strongly coupled to the vapor front, but after approximately 200 ns, it becomes detached. The position of the leading edge of the shockwave is somewhat challenging to precisely determine because of its non-hemispherical shape. The

Table 1
Matrix optical properties (266 nm).

	Molar extinction coefficient (l/mol cm)	Molarity (mol/l)	Absorption coefficient (cm^{-1})
Toluene	138 [24]	9.4	2987
Acetone	44.3 [24]	13.6	1388

anisotropic (R_z/R_x) nature of the expansion rises steeply at first and then levels off after 300–400 ns. This is similar, but somewhat different, to the behavior observed in Ref. [22], where a steep rise in the ellipticity of the shockwave was followed by a decrease towards a more hemispherical shape. Given the uncertainty in the position due to the challenge in determining the precise shape of the front, all further measurements of the shockwave position will be relative to the horizontal axis.

Typically, the position of laser-induced shock waves in air are fit to the Taylor–Sedov model [25] for a point blast,

$$R(t) = \alpha t^q \tag{1}$$

where α is a constant that depends on the energy of the blast wave and the thermodynamic properties of the medium, t is the time, and $q = (2/n+2)$ where n is the dimensionality. In the case of a laser-generated blast wave utilizing a circular beam in ambient