Photodeflection probing of the explosion of a liquid film in contact with a solid heated by pulsed excimer laser irradiation

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The explosion or rapid vaporization of a liquid film on an opaque surface by a pulsed laser is studied experimentally. Using a probe-beam deflection sensing (PDS) scheme, together with a previously developed transmission monitor, the distortion of the PDS signal due to the generation of shock waves by the exploding liquid is investigated. Various liquids, including alcohols and pure water in contact with substrates such as polyimide, amorphous carbon, and silicon, are studied for a wide range of excimer laser fluences. It is concluded that the present PDS technique is highly sensitive to the explosion threshold.

I. INTRODUCTION

Recently, there has been interest in the study of the explosion of a liquid film which is strongly superheated by a laser pulse on a solid substrate surface.[1–3] This is partly motivated by the desire to understand the mechanisms involved in the removal of particulates from contaminated surfaces by the explosive vaporization of a liquid film due to a pulsed laser irradiation.[4–6] Previously, we have developed piezoelectric[7] as well as optical transmission[1,3] techniques to probe the explosion. However, the piezoelectric transducer can only be positioned at a relatively large distance ($\gtrsim 1$ cm) from the liquid due to its relatively large size and hence the signal is susceptible to attenuation and dispersion. The optical transmission probe is limited to certain types of thin film samples which have a temperature dependent absorption coefficient at the probe wavelength. Hence shock waves generated by the explosive vaporization of liquids cannot be studied by the piezoelectric technique and samples of thickness greater than a few millimeters are difficult to study by the optical transmission technique. In the present work we have developed a more general probe-beam deflection sensing (PDS) technique to study the exploding liquid in close proximity ($\lesssim 1$ mm) to the substrate and for any kind of opaque substrate surface.

II. EXPERIMENTAL SETUP

Figure 1 shows the setup of our experiment. We have built a compact PDS sensing unit which contains a sample holder mounted on various X-Y-Z stages with the He-Ne probe beam (633 nm) located at an adjustable distance from the sample surface. The probe laser is mounted on one side of the unit. The position sensor, which is made with a high speed bicell photodiode, is mounted on the opposite side. The rise time of the bicell is about 10 ns. The He-Ne beam is expanded by a fivefold expander to about 2.5 mm in diameter and is focused by a lens with a focal length of 5 cm. The theoretical beam waist (w) at the focal spot is calculated to be about 16 μm assuming a Gaussian beam profile. A direct measurement gives a value for w of about 43 μm in air, with w expected to be larger in water.[8] The probe beam is about 0.4 mm from the surface of the sample. The KrF ($\lambda=248$ nm and 16 ns pulse width) UV excimer laser spot size was measured with thermal paper and found to be about 1 cm^2. A slit is used so that the width of the illuminated area of the sample is limited to 3 mm. This optimizes the PDS signal for the measured probe beam waist width.[9] The energy of the UV pulse is measured by a pyroelectric energy meter (Moletron JD 1000). By having the PDS experiment carried out in a compact unit such as the present one, interference from external vibrations is greatly minimized. In this experiment, we only report on the photoacoustic deflection signal[10] and ignore the photothermal signal[11] which is recorded at a delay time of a few μs after the UV pulse. In addition, for

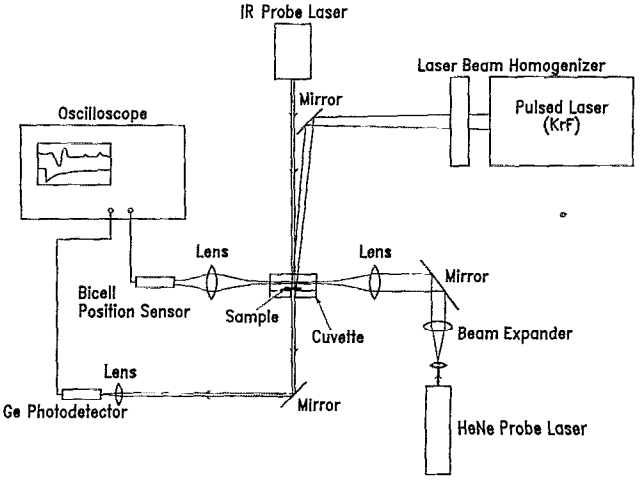

FIG. 1. Experimental arrangement for simultaneous transmission monitoring and photoacoustic probe-beam deflection sensing (PDS).

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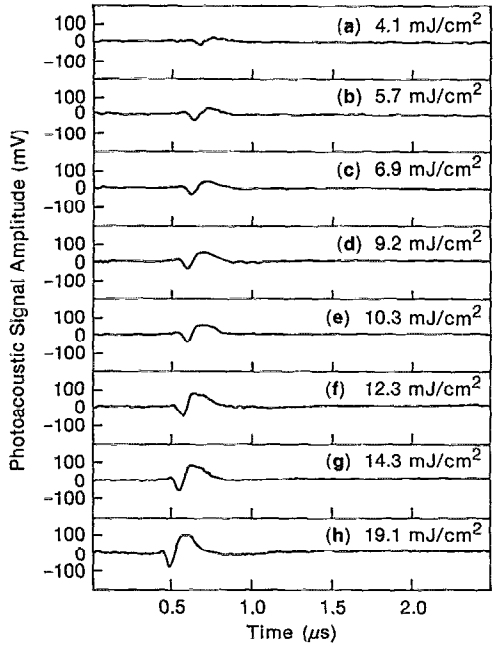

FIG. 2. Evolution of PDS signals with increasing KrF excimer laser fluence for the ablation of polyimide in air.

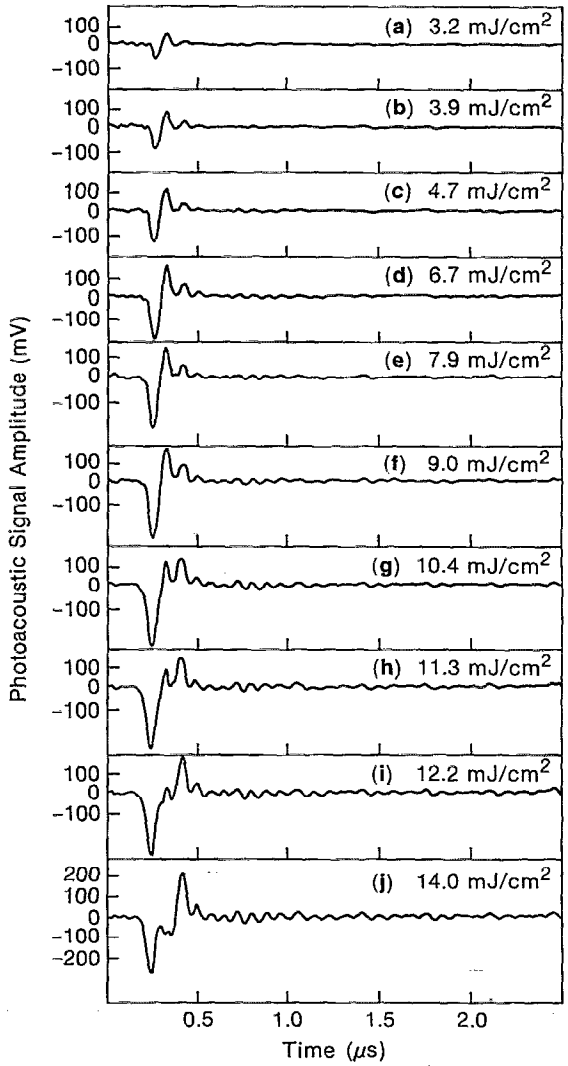

FIG. 3. Evolution of PDS signals with increasing excimer laser fluence for polyimide in water.

thin film samples such as amorphous silicon deposited on quartz, a cw diode (752 nm) laser probe was used for optical transmission measurements.[1,3]

III. RESULTS AND DISCUSSION

Figure 2 shows results for the ablation of polyimide (PI) in air. The evolution of the PDS (photoacoustic) signal with increasing laser fluence agrees with recent results published for polyethylene terephthalate (PET) ablation in air.[12] With increasing fluence, the signal becomes more asymmetrical with the leading edge becoming sharper and the arrival time (measured from the end of the pulse) becoming shorter. These features can be explained by the production of a shock wave when ablation occurs. The signal is due to the superposition of an acoustic pulse produced by the so called "thermal piston,"[7] and a compressional shock of single polarity. The ablation threshold defined by the change of the shape of the signal is about 10 mJ/cm^2 for PI. This is lower than the 17 mJ/cm^2 threshold found previously using a piezoelectric probe.[7] This suggests that the PDS technique is more sensitive to the onset of ablation, in agreement with a recent study comparing the sensitivity of the PDS technique with optical reflection probes for monitoring the threshold of pulsed laser damage of surfaces.[13]

To study the explosion of strongly superheated water at the PI/water interface, we put the PI film in a quartz cuvette which is filled with water. The excimer pulse transmits through the quartz window and water and is absorbed by the PI. Heat then diffuses into the water and superheats it to a temperature well above the normal boiling temperature. The motivation for this study comes from our desire to understand the basic mechanism involved in the "steam

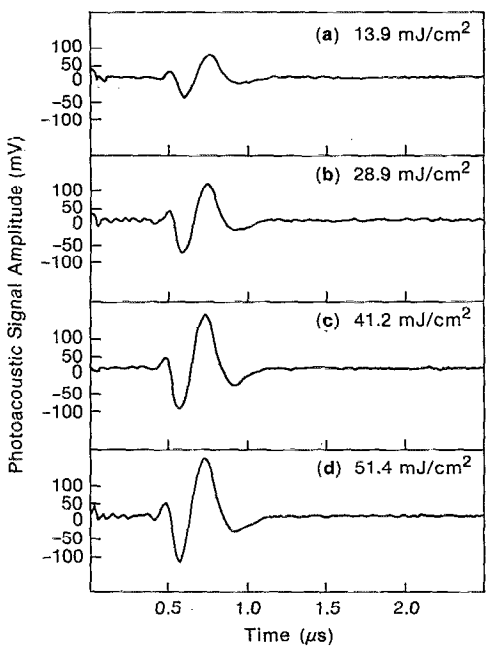

FIG. 4. PDS signals for amorphous carbon (a-C) in air at various KrF excimer laser fluence.

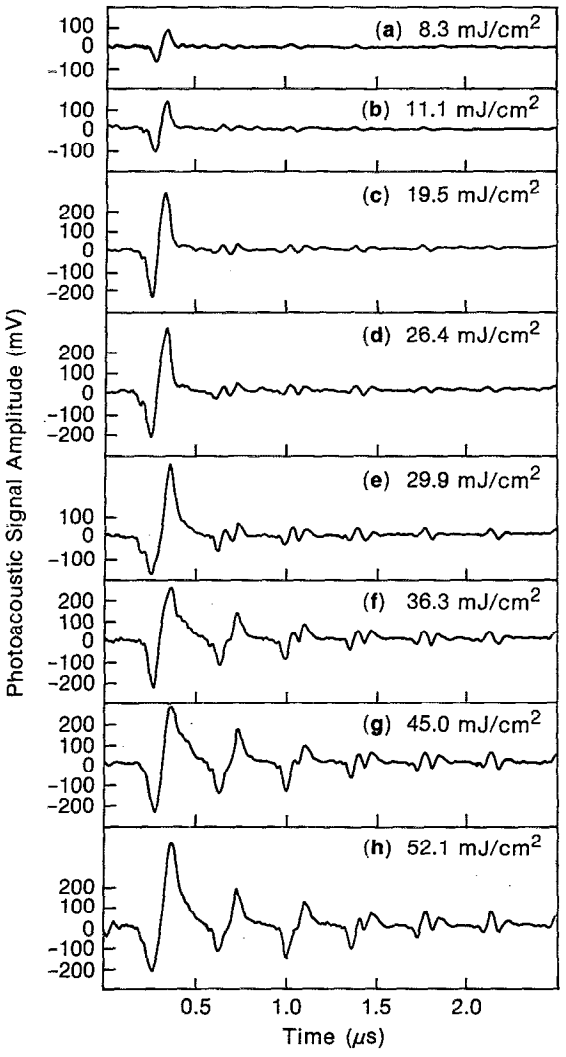

FIG. 5. PDS signal for *a*-C in water with increasing excimer laser fluence.

laser cleaning" technique recently developed by us,[4,6] in which a micron thick liquid film is deposited on a contaminated surface and is exploded away by an excimer laser pulse. Particulates are removed by the pressure generated

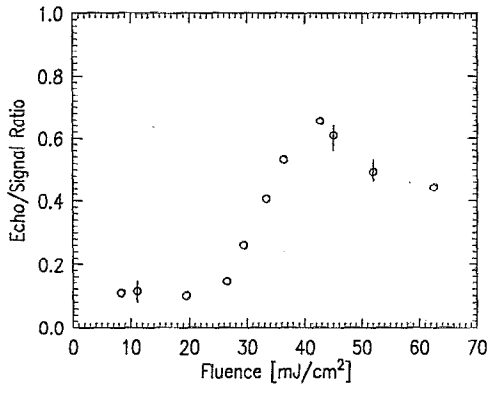

FIG. 6. Magnitude of the first echo from the multiple reflections (normalized by the magnitude of the first pulse in the PDS signal) vs fluence for *a*-C in water.

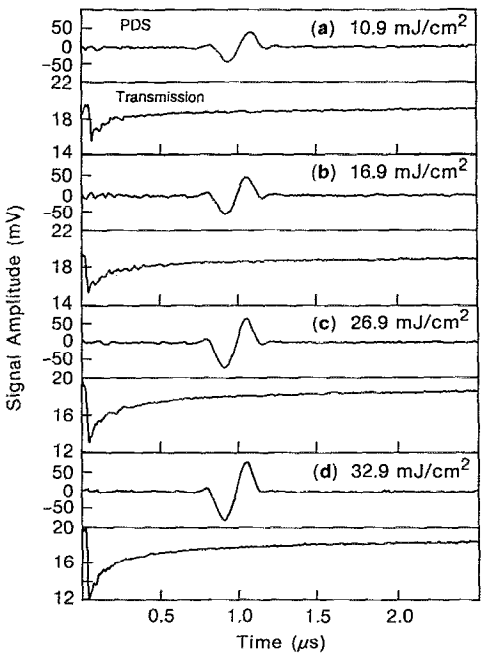

FIG. 7. PDS and transmission signals for *a*-Si in air with increasing excimer laser fluence.

by the exploding liquid film. It is of interest to note that a similar experiment has been carried out recently[14] to study the shock wave generated by the pulse ablation of PI in water. However, that experiment was done at much higher laser fluences which generated shocks of much higher pressure (~kbar). Here, we "explode" the water at the interface using a much lower range of fluences for which we expect the shocks generated by the explosion of superheated water to have a pressure up to ~100 bars.[15] Figure 3 shows the PDS signals for various laser fluences. We note that the signal becomes distorted at about 10 mJ/cm^2. In addition, above 10 mJ/cm^2 the echoes reflected from the PI (0.07-mm-thick)-glass (1-mm-thick) interface and the back surface of the glass substrate are strong. This indicates that the temperature at the water-PI interface is high enough to superheat the water to explosion, creating shock waves and hence strong PDS signals.

Figure 4 shows the PDS signals for a thick (0.8 mm) high-density amorphous carbon (*a*-C) sample in air at different fluences. There is no observable ablation of the sample up to about 50 mJ/cm^2 as indicated by the constant shape of the PDS signal with increasing fluence. Figure 5 shows data obtained with the same *a*-C sample in the water

TABLE I. Threshold fluences for the explosion of different liquids on different substrates.

	Liquids			
Substrates	H_2O	Iso-propanol	Ethanol	Methanol
a-Si	20 mJ/cm^2	10 mJ/cm^2	10 mJ/cm^2	9 mJ/cm^2
a-C	26 mJ/cm^2	17 mJ/cm^2	17 mJ/cm^2	15 mJ/cm^2
Polyimide	10 mJ/cm^2	9 mJ/cm^2	9 mJ/cm^2	7 mJ/cm^2

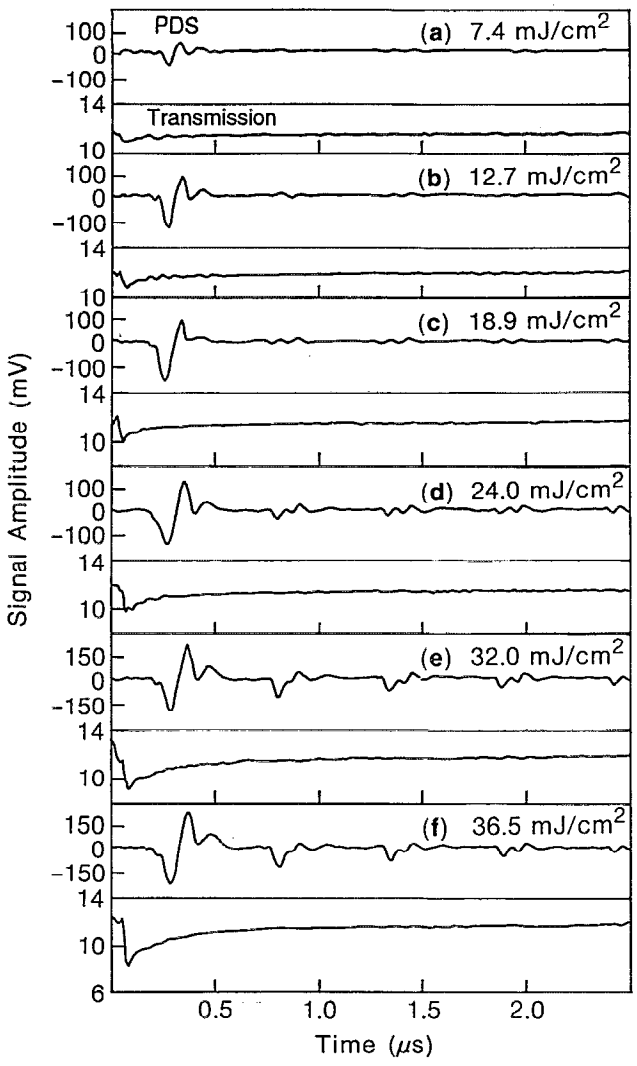

FIG. 8. PDS and transmission signals for a-Si in water with increasing excimer laser fluence.

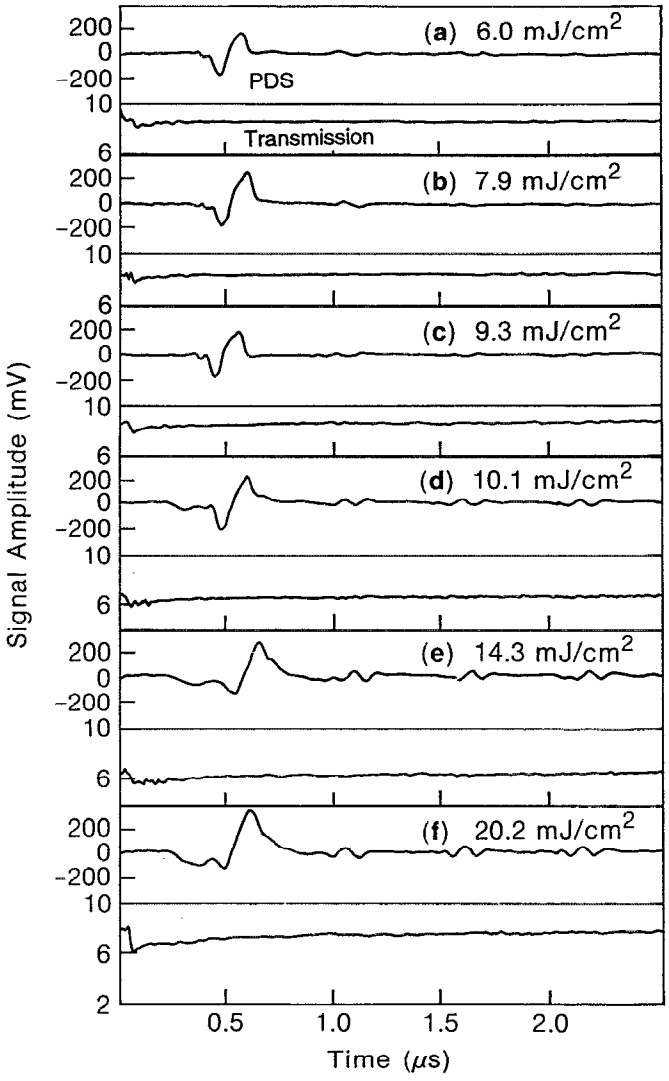

FIG. 9. PDS and transmission signals for a-Si in iso-propanol with increasing excimer laser fluence.

cuvette. Since the sample does not ablate within this range of fluences, we attribute the change in the PDS acoustic signal to the explosion of the liquid. The threshold for this to occur is indicated by the presence of strong echo pulses reflected from the back surface for laser fluence greater than 26 mJ/cm^2. We find this is the better way of defining the threshold as compared to the case of ablation of PI in air, in which the change of the pulse shape is used. We have plotted in Fig. 6 the ratio of the amplitude of the first echo (normalized to the initial signal amplitude) for increasing laser fluence. The jump in the plot at a fluence of 26 mJ/cm^2 indicates the threshold for the explosion of the liquid. We believe that below the explosion threshold, thermoelastic expansion is the only mechanism producing the observed acoustic pulse. With our present UV beam size, the wave generated in a solid sample is primarily a shear wave. Such generated shear waves do not convert into longitudinal waves on normal reflection. Therefore, there is no longitudinal pulse re-entering the water at the water-sample interface and hence little PDS echo signal is detectable. Above the explosion threshold, there is a strong longitudinal pulse produced by the "explosion recoil" at the interface. This longitudinal pulse undergoes multiple reflections within the sample, and produces multiple pulses in water, giving multiple PDS signals. We have also obtained data with other liquids including different kinds of alcohols. The explosion thresholds, as determined by the onset of multiple echoes for each liquid, are given in Table I. As expected, the thresholds for alcohols are lower than those for water.[3]

We have also carried out a similar experiment on a 0.2-μm-thick amorphous silicon (a-Si) film deposited on a 1.5-mm-thick fused quartz substrate. Beside looking at the PDS signal, we also examined the transmitted signal probed by a cw diode laser at 752 nm. Since the absorption coefficient at 752 nm increases with temperature, the magnitude of the transmission signal is a measure of the average temperature of the a-Si film.[3,16] Figure 7 shows the PDS and IR transmission signals when the a-Si film is in air. No ablation is indicated in this range of fluence. Figures 8 and 9 show the signals when the a-Si is in water and

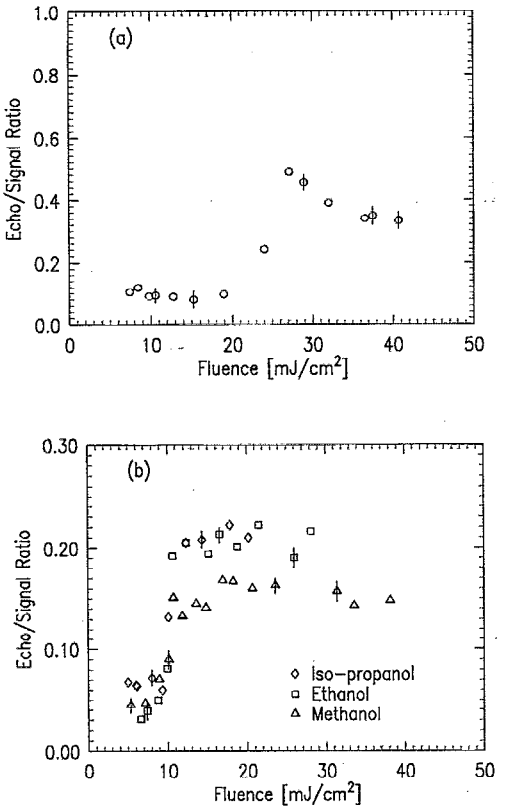

FIG. 10. Magnitude of the first echoes normalized to the initial signal vs fluence for (a) *a*-Si in water and in (b) alcohols.

in iso-propanol, respectively. From the magnitude of the echoes reflected within the sample, the explosion thresholds are found to be 20 mJ/cm^2 for water and 10 mJ/cm^2 for iso-propanol. The explosion threshold for other liquids is also listed in Table I. Figure 10(a) plots the magnitude of the first echo normalized to the initial signal versus fluence for the *a*-Si sample in water and Fig. 10(b) plots that for alcohols. In these figures, a sudden change of slope which corresponds to the liquid explosion at the interface is observed. This is consistent with our observation for *a*-C samples.

IV. CONCLUSION

In conclusion, we have shown in this article that the PDS technique is useful for monitoring the explosion of liquids when irradiated by excimer laser pulses. When the liquid at the interface explodes, longitudinal echoes are generated due to the multiple reflection in the solid sample. We find that the sudden increase in longitudinal echo magnitude is a reliable indication of the occurrence of explosion at the interface. We find that the PDS probe is a useful technique to monitor the longitudinal echoes that couple into the liquid.

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