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Ultrafast stress pulse detection by laser beam deflection

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Abstract: We describe a direct method for the detection of optically excited ultrashort stress pulses in thin films using a time-resolved pump and probe scheme. Changes in the surface profile are monitored by the angular deflection of a probe beam. Application to the detection of interfacial layers is also demonstrated.

The time-resolved detection of high frequency phonons excited by picosecond or femtosecond optical pulses can be achieved by a variety of methods. Terahertz optical phonons, for example, have been detected through the electrooptic effect,[1] whereas acoustic phonons up to the 100-GHz range have been detected through the photoelastic effect.[2] Here we describe an ultrafast method for the detection of stress pulses associated with longitudinal acoustic phonons, which is based on laser beam deflection from ultrafast surface vibrations.[3] Such a detection scheme has been widely used by the photoacoustics community in lower frequency photothermal deflection or photothermal displacement experiments.[4, 5]

Optical pump pulses of duration 3 ps (FWHM), repetition rate 76 MHz, wavelength $\lambda = 630$ nm and energy 0.6 nJ are used to excite the stress pulses. Light absorbed within the optical absorption depth $\xi$ ($\sim 10$ nm for the metal samples used here) is converted to heat, and, through the resulting thermal expansion, longitudinal stress pulses in the 100-GHz region are generated. Changes in the surface slope of the opaque films, induced by the stress pulses bouncing back and forth inside the films, are interrogated by measuring the angular deflection ($\theta \sim 1$ grad) of a probe laser beam derived from the same laser (see Fig. 1). This beam is focused to a 20-µm diam. spot size to partially overlap with the pump spot of similar size. The angular deflection is monitored with a dual-element photodiode. The displacement of the surface, typically $\sim 0.001$ nm, can therefore be measured. By scanning the delay line, time-resolved detection on a picosecond time scale is achieved. This angular deflection scheme was previously used to measure the transient thermal expansion of bulk crystalline silicon at lower frequencies of order 2 GHz, but not for the detection of stress pulses.[6]

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Fig. 1: Results using the beam deflection detection method: probe beam angular deflection ($\delta \vartheta$) against delay time for a 480-nm Al film on a sapphire substrate. Lower curve: theoretical fit to echoes with a thermoelastic model. Square inset: theoretical stress pulse shape (on the same time scale). Circular inset: experimental geometry—the solid ray represents the probe beam.

Fig. 2: Results using the reflectance detection method: relative reflectance variation for the 480-nm aluminium film of Fig. 1. Lower curve: theoretical fit. The relative intensity change $\delta I/I$ for the first echo is about $4 \times 10^{-6}$, where $I$ is the probe beam intensity.

The results for a sputtered aluminium film of thickness 480 nm on a sapphire substrate are shown in Fig. 1. Echoes are clearly visible. These arise primarily because of the surface displacement, although we also expect a contribution from near-surface radial gradients in the dielectric constants associated with the time-varying lateral stress distribution.[3] For Al we estimate that the displacement contribution is dominant, and the fit in Fig. 1 represents this contribution based on a thermoelastic model with lossless, nondispersive stress propagation. Diffusion processes and frequency-dependent ultrasonic attenuation are responsible for the observed stress pulse broadening. Extracting the stress pulse shape, proportional to the time derivative of the displacement contribution,[3] allows
the stress pulse broadening to be probed quantitatively. (This can be easily done for materials in which the contribution from near-surface radial gradients is relatively small.\[^{[3, 7]}\]) The negative step in Fig. 1 is caused by nonequilibrium electron heating.\[^{[8]}\] The sign of the signal changed, as expected, when the probe beam was focused on the opposite side of the pump beam spot.

For comparison, results using the time-resolved reflectance detection method\[^{[1]}\], based on the photoelastic effect, are shown in Fig. 2 for the same sample. The step near delay time \(t=0\) and overall background decay are caused by the initial temperature change and subsequent thermal diffusion. The echo shape depends in a complex way on the optical properties of the material \((n\) and \(k)\) and on its (often unknown) photoelastic constants \(\frac{dn}{d\eta} = \frac{dk}{d\eta}, \eta\) the strain). Quantitative investigations with this method are therefore difficult. In absolute terms the relative change in intensity \(\frac{\delta I}{I}\) \((I\) the probe beam intensity) for the echoes is about 7 times lower than that for the angular beam deflection results \(\frac{(I_1-I_2)}{I_1}\), where 1 and 2 correspond to the two elements of the dual-element photodiode. However, beam pointing fluctuations contribute to the noise in the measurements of \(\delta I\), and so the overall signal-to-noise ratio is similar in Figs. 1 and 2. With active beam pointing stabilization it may be possible to obtain a better resolution for Al films with the beam deflection detection. In other thin film samples such as Au, Ag or a-Si, echoes could only be resolved with the beam deflection method.\[^{[7, 9]}\]

Thin interfacial impurity layers can significantly affect the film adhesion. We therefore investigated a sample with such a layer. Results are shown in Fig. 3 for a multilayer structure containing a Cr/W interface with a thin interfacial amorphous silicon layer. The Cr and W were prepared by sputtering as films of thickness \(\sim 600\) nm on a Cr substrate. Samples with 50- and 10-nm sputtered a-Si layers were produced. The presence of the a-Si layer changes the first echo shapes owing to multiple acoustic reflections inside this layer (which has an acoustic impedance much less than that of Cr or W). The stress pulse shapes based on thermoelastic stress generation are shown in the insets, and the echo shapes derived (from the surface displacement variation—contributions from radial gradients of the dielectric constants are small\[^{[3]}\]) are also shown in Fig. 3 (including the effect of multiple acoustic reflections up to order 7). The reasonable agreement with experiment indicates good bonding between the different layers. Similar detection of interfacial layers with thicknesses down to 0.1-nm order has previously been demonstrated.\[^{[3, 10, 11]}\]

We have also successfully applied this laser beam deflection method to films of Mo, W, Ni, Nb, V, Ta, Cu, Pt and a-Ge. In contrast to the reflectance detection method, for which the effects of optical penetration limit the detection bandwidth to \(\sim 100\) GHz, the beam deflection detection method, essentially local to the surface, is only limited by the optical pulse duration. For a Gaussian optical pulse of FWHM duration \(\tau\), the frequency for signal reduction by a factor of 2 (compared to \(f=0\)) is \(f\approx 0.44/\tau\approx 200\) GHz for the present experiments. Entering the realm of THz laser ultrasonics should therefore be possible using shorter optical pulses on the order of 200 fs or less, providing that high frequency acoustic
Angular beam deflection against delay time for two samples containing a Cr/a-Si/W interface with a-Si thickness a) 50 nm and b) 10 nm. The background variation has been subtracted. The upper curves are theoretical fits according to the thermoelastic model. The insets show the calculated stress pulse shapes (on the same time scale).

generation can be achieved.

In conclusion, we have presented a non-contact laser picosecond acoustic method based on laser beam deflection for application to thin opaque films and nanostructures. Quantitative investigation of the mechanisms of picosecond laser-induced stress generation or evaluation of film or interfacial layer thicknesses and their bonding is possible.