Picosecond Dynamics of Thermal and Acoustic Transport in Metal Films[1]

G. L. Eesley[2]

An optical transient reflectance technique has been developed to characterize both the thermal and the elastic properties of thin-film materials. The use of picosecond-duration laser pulses permits the study of metal films of the order of 100 nm thick. We demonstrate the measurement of thermal diffusivity and acoustic velocity, in addition to observations of the thermal impedance of a single metal–metal interface.

KEY WORDS: high-speed techniques; reflectance; sound velocity; thermal diffusivity; thin films.

1. INTRODUCTION

Increasing interest in new thin-film materials has motivated the development of characterization techniques which are capable of measuring film transport properties independent of the supporting substrate. To supplement our materials research involving compositionally modulated metal films, we have developed a transient reflectance technique which is capable of providing information on both the thermal and the elastic properties of supported thin metallic films. We can measure the thermal diffusivity of films on the order of 100 nm thick, independent of the substrate thermal properties. Our measurement is also sensitive to the impedance of thermal transport by a single metal–metal interface. In addition, we can generate and monitor the propagation of picosecond-duration acoustic pulses in thin metal films and this permits a determination of the film elastic response.

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2. TECHNIQUE AND APPARATUS

The transient reflectance measurement utilizes two synchronous picosecond-laser pulses to heat the sample surface and to probe the time response of the reflectivity change produced by the ultrafast heating. For small temperature deviations, the change in reflectivity will be linearly related to the surface temperature increase [1, 2]. In the case of metals, the reflectivity change is due largely to an increase in electron–phonon collisions. Typical fractional changes of $\Delta R/R \sim 10^{-5}$ can be expected for temperature changes of the order of 10 K. Since the penetration depth of visible light (and thus the heating depth) is of the order of 20 nm, thermal diffusion out of this region will occur within a few hundred picoseconds. Therefore by monitoring the transient reflectivity change on this time scale, we complete our measurement before substrate effects become important for films > 100 nm thick.

The thermal expansion which accompanies the surface heating results in the generation of a longitudinal acoustic pulse which propagates away from the surface [3]. This pulse will be partially reflected by the film–substrate boundary and return to the surface. In addition to the thermally induced reflectivity change, the returning acoustic pulse will produce an expansion-induced reflectivity change with duration determined by the transit time of the acoustic pulse within the optical skin depth. It is the time-resolved measurement of these thermoreflectance and piezoreflectance signals which forms the basis of our technique. Although our studies have
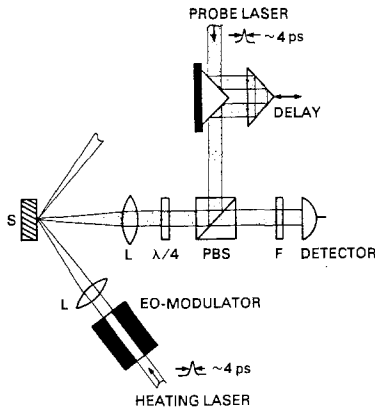
Fig. 1. Schematic of the transient reflectance optical arrangement. L, lens; F, filter; PBS, polarization beamsplitter; $\lambda/4$, quarter-wave plate.

concentrated on novel metallic films, extension of our measurements to thin film dielectrics and semiconductors is possible [4].

The apparatus consists of two synchronously pumped dye lasers which generate 4-ps pulses at a wavelength of 633 and 595 nm for the heating and probing lasers, respectively. The average heating laser power at the sample is ∼45 mW and the probing power is ∼2 mW. The probing pulse can be time delayed relative to the heating pulse, and zero time delay is determined by a cross-correlation measurement of the two pulses. Lockin detection techniques are used to measure the changes in surface reflectivity as a function of time delay between the amplitude-modulated heating pulse train and the probing pulse train. Figure 1 is a schematic of the optical configuration we use to monitor the changes in reflectivity of the sample surface. The details of the apparatus and data collection procedures are given in Ref. 2.

3. THERMOREFLECTANCE

We have used transient thermoreflectance (TTR) to measure the thermal diffusivity of both evaporated and ion sputter-deposited single-element and compositionally modulated metal films [2, 5]. Because the heating occurs within a volume roughly 20 μm in diameter and 20 nm thick, the diffusion of heat out of this region can be described by a one-dimensional heat flow equation. The solutions of this equation are iteratively fit to the TTR signal with the thermal diffusivity as a free parameter. We assume that the TTR signal corresponds to the surface temperature of the sample, and we neglect reflectance signals which may originate from below the metal surface. This procedure may lead to error in the determination of absolute thermal diffusivity values. The conditions under which volume contributions to the TTR signal are important remain unclear at present and further experimental work to delineate the details of volume reflectance is needed [6].

Nonetheless, we do account for volume heating in the sample and solutions of the heat flow model accurately predict the temporal response of the surface temperature in the presence of an extended temperature gradient below the surface. TTR observations can be used to observe the relative changes in thermal transport which may accompany variations in material structure and elemental composition.

A rather dramatic demonstration of the sensitivity of thermal transport to the presence of a metal–metal interface is shown in Fig. 1. The TTR signals from single-element Ni and Ti films and a Ni/Ti bilayer film are shown in Fig. 2, and the geometry of the single-element and bilayer films is represented in Fig. 3. The films were magnetron sputter deposited to a
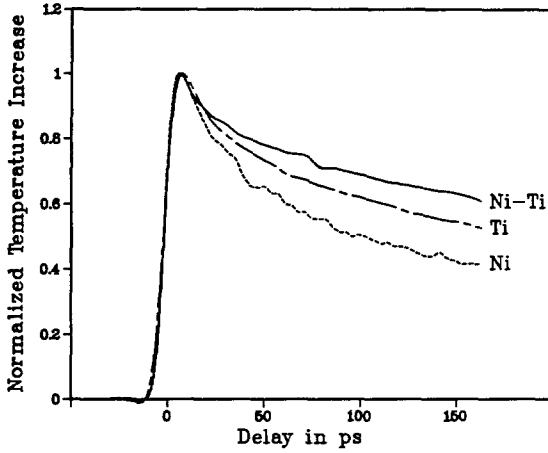
Fig. 2.   TTR signal from the Ni film, the Ti film, and a Ni/Ti
bilayer film.

thickness of 300 nm in the case of the Ni and Ti films, and the bilayer film
is composed of a 300-nm Ti underlayer with a 30-nm Ni overlayer. All films
were deposited onto an oxidized silicon wafer.

In Fig. 2 we see that the heat deposited at zero time delay diffuses
away from the surface region faster in Ni than in Ti, as would be expected
from the relative diffusivities of these two materials. We determine values
of $4.4 \times 10^{-6} \, m^2 \cdot s^{-1}$ and $1.4 \times 10^{-6} \, m^2 \cdot s^{-1}$ for the Ni and Ti films,
respectively. We see from the bilayer signal that the presence of a single
interface between Ni and Ti seriously degrades the thermal transport
relative to that expected from the presence of a thick Ti underlayer.

We have analyzed this measurement as well as other TTR data from
a series of metal–metal pairs in both bilayer and multilayer structures. The
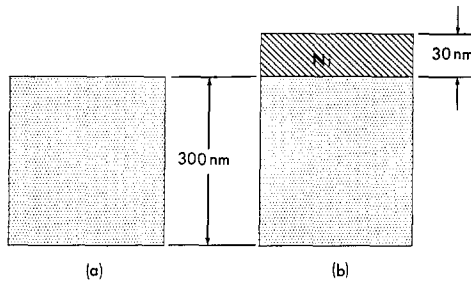results are described in detail in Ref. 5.

Fig. 3.   Geometry of the sample films used for the
measurements shown in Fig. 2: (a) single-element film
and (b) bilayer film.

4. PIEZOREFLECTANCE

In conjuction with our TTR studies of thin-film thermal transport, we have generated and observed picosecond-duration acoustic pulses in a number of single-element and compositionally modulated metal films. The acoustic pulses are generated by the thermoelastic deformation of a free surface heated by a short optical pulse [7, 8]. The optical energy absorbed in the illuminated volume results in local heating and thermal expansion of the surface region and a longitudinal acoustic pulse is launched into the film. Since the material compression and tension are in the direction of propagation, the pulse travels with a velocity appropriate to a conventional longitudinal acoustic wave. The velocity is related to the longitudinal elastic constant ε by the relation $v = \sqrt{(\varepsilon/\rho)}$, where ρ is the material density.

The acoustic pulse propagates into the film and a fraction of the pulse is reflected by abrupt gradients in density or elastic properties. In the case of a thin supported film, the portion of the pulse reflected at the film–substrate interface will return to the surface and it can be detected as a change in reflectivity. The mechanism of the reflectivity change is the alteration of band structure which results from the compression of the metal lattice.
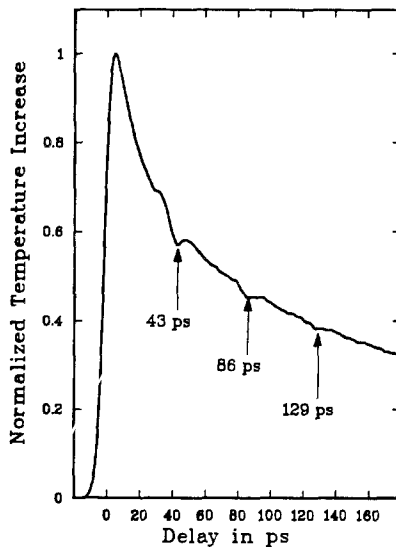

Fig. 4. Thermoreflectance and piezoreflectance signals from a 125-nm-thick Ni film. The acoustic pulse-echoes are indicated by arrows.

The transient piezoreflectance (TPR) measurement simply determines the time delay associated with an acoustic pulse which has traversed the sample film, and knowing the deposited thickness, we can calculate the sound velocity. Thus the measurement is an ultrafast pulse-echo technique which is fully analogous to conventional ultrasonic measurements. The primary advantage of the TPR measurement is the ability to measure picosecond time delays in thin films, without contacting the film with transducers.

To demonstrate this technique, we have performed measurements on a sputter-deposited 125-nm Ni film supported by an oxidized silicon wafer. Figure 4 shows the superimposed TTR/TPR signal normalized to the peak TTR signal at zero time delay. An acoustic pulse which has completed one round trip in the film is visible as a dip in the TTR profile at a delay of 43 ps. Multiple reflections at 86 and 129 ps are also seen and these correspond to pulses which have completed two and three round trips in the film, respectively. The detected profile of the acoustic pulse will be complicated by detection of TPR signals which originate from within the optical skin depth, as well as the fact that the compression/expansion profile of the pulse reflects back onto itself at the sample surface. In general, a bipolar signal profile is not expected in a TPR measurement. The slight increase in signal on the leading and trailing edge of the acoustic pulses in Fig. 4 results from these considerations [4].

The overall decay profile of the TTR contribution to the data in Fig. 4 can be fit by our heat flow model and subtracted from the signal to retrieve the TPR signal alone. This result is shown in Fig. 5, where the peak frac-
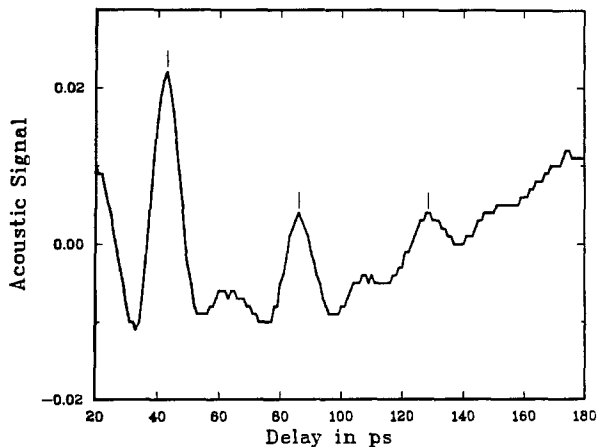

Fig. 5. Transient piezoreflectance contribution to the signal in Fig. 4.

tional change in reflectivity produced by the first pulse-echo is $\Delta R/R = 1.2 \times 10^{-6}$. Although this is an extremely small signal, the experimental configuration we use can easily detect these changes. The variation in the nonzero background signal in Fig. 5 results from slight fitting differences between the TTR data and the one-dimensional model calculation. These differences arise from noise in the data and fitting error introduced by the presence of several closely spaced acoustic echoes which are not accounted for by the heat flow model.

The measured time delay of 43 ps and film thickness of 125 nm yield a sound velocity of $5814\ \mathrm{m \cdot s^{-1}}$. This compares with literature values of 5300, 6000, and $6200\ \mathrm{m \cdot s^{-1}}$ for magnetically saturated Ni in the (100), (110), and (111) crystal directions, respectively [9]. We have not investigated the magnetic state, domain structure, or orientation of this particular sample. Repeated measurements on this sample would indicate a $\pm 5\%$ accuracy in determining the sound velocity.

5. CONCLUSIONS

In conclusion, we have demonstrated a time-resolved measurement of both the thermal and acoustic transport dynamics in thin, supported metal films. Our transient thermoreflectance and piezoreflectance measurements can be extended into the femtosecond time domain to investigate the dynamics of energy transport under nonequilibrium electron-lattice temperature differences [10]. The technique is noncontacting and substrate independent, and we expect that this will be useful for characterizing thermal transport at metal–metal and metal–semiconductor contacts. We are currently applying the piezoreflectance technique to studies of the changes in elastic properties of compositionally modulated metals as a function of the composition repeat distance. Extension of this measurement technique to other materials should be straightforward.

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