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Thin-Film Thermophysical Property Characterization by Scanning Laser Thermoelectric Microscope[1]

T. Borca-Tasciuc[2] and G. Chen[2, 3]

This work presents a scanning laser-based thermal diffusivity measurement technique for thin films as well as for bulk materials. In this technique, a modulated laser beam is focused through a transparent substrate onto the film–substrate interface. The generated thermal wave is detected using a fast-responding thermocouple formed between the sample surface and the tip of a sharp probe. By scanning the laser beam around the thermocouple, the amplitude and phase distributions of the thermal wave are obtained with micrometer resolution. The thermal diffusivity of the film is determined by fitting the obtained phase signal with a three-dimensional heat conduction model. Experimental results are presented for a 150-nm gold film evaporated on a glass substrate.

KEY WORDS: photothermal method; scanning laser; thermal conductivity; thermal diffusivity; thermoelectric effect; thin film.

1. INTRODUCTION

Measurement of the thermophysical properties of thin films is a challenging task, and many techniques have been developed. Available techniques and their range of applications have been summarized in several publications [1–4]. To determine the in-plane thermal conductivity or thermal diffusivity of thin films, substrate removal is often necessary. Alternative thin-film thermophysical property measurement techniques, particularly methods that are capable of determining anisotropic thermophysical properties of thin films without removing the substrate, will provide valuable tools for

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[2] Mechanical and Aerospace Engineering Department, University of California at Los Angeles, Los Angeles, California 90095, U.S.A.

[3] To whom correspondence should be addressed.

understanding heat transfer in thin films and for exploring thin film based devices.

The difficulty in determining the in-plane thermophysical properties of thin films lies in the fact that most heat is conducted through the substrate rather than through the film. One possible way to overcome this difficulty is to obtain the two dimensional temperature distribution around a microscale heat source. From the spread of the heat source, it may be possible to obtain anisotropic thermophysical properties of the thin film and/or the bulk material. One technique that gives the two-dimensional thermal field of a thin film on substrate structure is an optical pump-and-probe method by scanning either the pump laser or the probe laser [5–7]. The disadvantage of this method is the difficulty of aligning two laser beams through the focusing optics. Kemp et al. [8] developed a method that maps the temperature distribution of a free-standing film generated by a scanning laser and detected by a thermocouple formed by welding a fine bismuth–antimony–telluride probe to the sample.

This paper extends the idea of Kemp et al. [8] for free-standing thin films and develops a method suitable for determining anisotropic thermophysical properties of thin films for film-on-substrate systems without the removal of the substrate. The method employs modulated laser radiation to generate a thermal wave in the sample and scans the focused laser beam over the area of interest. The laser wavelength is tuned to a range where the substrate is transparent such that the laser beam penetrates the substrate and acts as a micrometer scale heat source in the film. The detection is done with a fast thermoelectric effect created by direct contact between the "metallically" covered surface of the sample and the sharp tip of an electrochemically etched wire. Thermophysical properties of the film or the substrate material can be determined from the amplitude and/or the phase of the thermoelectric voltage signal.

2. EXPERIMENTAL

The method is designed to measure thermophysical properties of films on a substrate or bulk materials. A focused laser beam is scanned through the transparent substrate over the film–substrate interface in order to produce thermal waves that are detected using a fine thermocouple formed between the sample and the probe. Thermophysical properties can be determined from the amplitude and/or phase profiles of the data. Our current experimental setup is shown in Fig. 1.[4] An argon laser is used to pump a tunable Ti–sapphire laser that can output laser radiation with a

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[4] A diode laser is preferable to the Ti–sapphire laser for a compact system.

wavelength between 700 and 900 nm. The modulated radiation is coupled into a single-mode optical fiber. The optical system used to focus the laser radiation is assembled onto a base plate, which is placed onto an *x–y* scanner (in our case, it is fitted into the scanner of an atomic force microscope). A specially designed holder is used to maintain the sample at an adjustable distance above the scanning laser such that radiation penetrates the transparent sample and is incident on the top surface. The top surface is a conducting element that absorbs the laser radiation and also forms one leg of the thermocouple. The other leg of the thermocouple is formed by a fine wire contacting the surface.

A series of experiments is conducted with a 150-nm gold film evaporated on the top surface of a glass slide. The probe used to detect the thermal wave is a sharp K-type thermocouple wire (nickel–aluminum) obtained through electrochemical etching in a solution of sulfuric acid. The initial diameter of the wire before etching is 25 μm. The tip of the etched wire is brought into contact with the gold film. The electrical contact resistance between the film and the tip may be used to estimate the contact radius [9]. The obtained value of ~180 Å is a measure of the spatial resolution of the probe in a vacuum environment. However, air conduction and the adsorbed water film that forms on the surface of the sample may

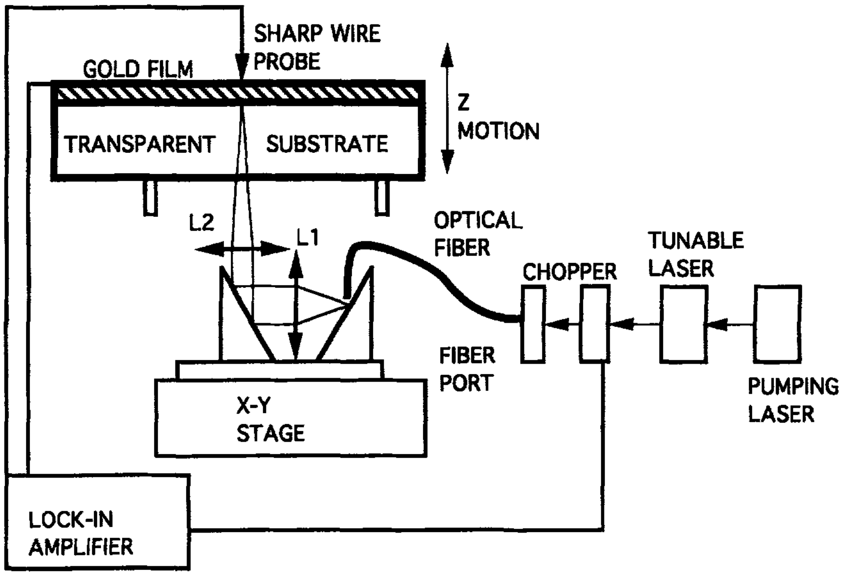


Fig. 1. Schematic diagram of the experimental setup.

reduce this resolution. The temporal resolution of the probe is related to the mass and heat capacity of the junction. Because the junction has practically zero mass, the thermocouple is expected to be very fast-responding, limited only by the electron response time. In this work, a mechanical chopper with a maximum frequency of 3.2 kHz is employed. The response time of the thermocouple cannot be determined. The AC components of the amplitude and the phase of the thermoelectric voltage developed at the tip–surface junction are picked up by a lock-in amplifier.

The laser was scanned over a 128×128-μm area around the probe. The minimum laser beam radius as estimated using the ABCD law is about 1 μm [10]. At a scan size of 128 μm and a round-trip frequency of 0.1 Hz (the minimum attainable frequency in the current setup), the scanning speed of the laser is $u = 25.6 \times 10^{-6}\ \mathrm{m \cdot s^{-1}}$. The maximum modulation frequency of our current setup is 3200 Hz. Under these conditions, the AC temperature detected by the stationary probe as a function of the laser beam location is equivalent to the AC temperature profile around a fixed heat source [11]. For a film-on-substrate structure being heated at the film–substrate interface, assuming that the film is optically thick, the AC temperature rise on the surface is given by the following expression, which contains both the amplitude and the phase information of the temperature profile [11]:

$$\theta_{\mathrm{fAC}}(r) = \frac{I_0 R^2}{2} \int_0^\infty \frac{\beta J_0(\beta r)\, e^{-R^2\beta^2/4} d\beta}{Z_\mathrm{f} \sinh(m_\mathrm{f} d) + Z_\mathrm{s} \cosh(m_\mathrm{f} d)} \tag{1}$$

where θ_{fAC} is the AC temperature rise on the film surface, r the radial coordinate, I_0 the absorbed laser beam intensity, R the radius of the laser beam, β the integration variable, J_0 the Bessel function of order zero, and d the film thickness. Other quantities in Eq. (1) are defined as

$$Z_\mathrm{s} = k_\mathrm{s} m_\mathrm{s}, \qquad Z_\mathrm{f} = k_{\mathrm{fz}} m_\mathrm{f} \tag{2}$$

$$m_\mathrm{s} = \sqrt{\beta^2 - i\omega/\alpha_\mathrm{s}} \tag{3}$$

$$m_\mathrm{f} = \sqrt{\gamma\beta^2 - i\omega/\alpha_{\mathrm{fz}}} \tag{4}$$

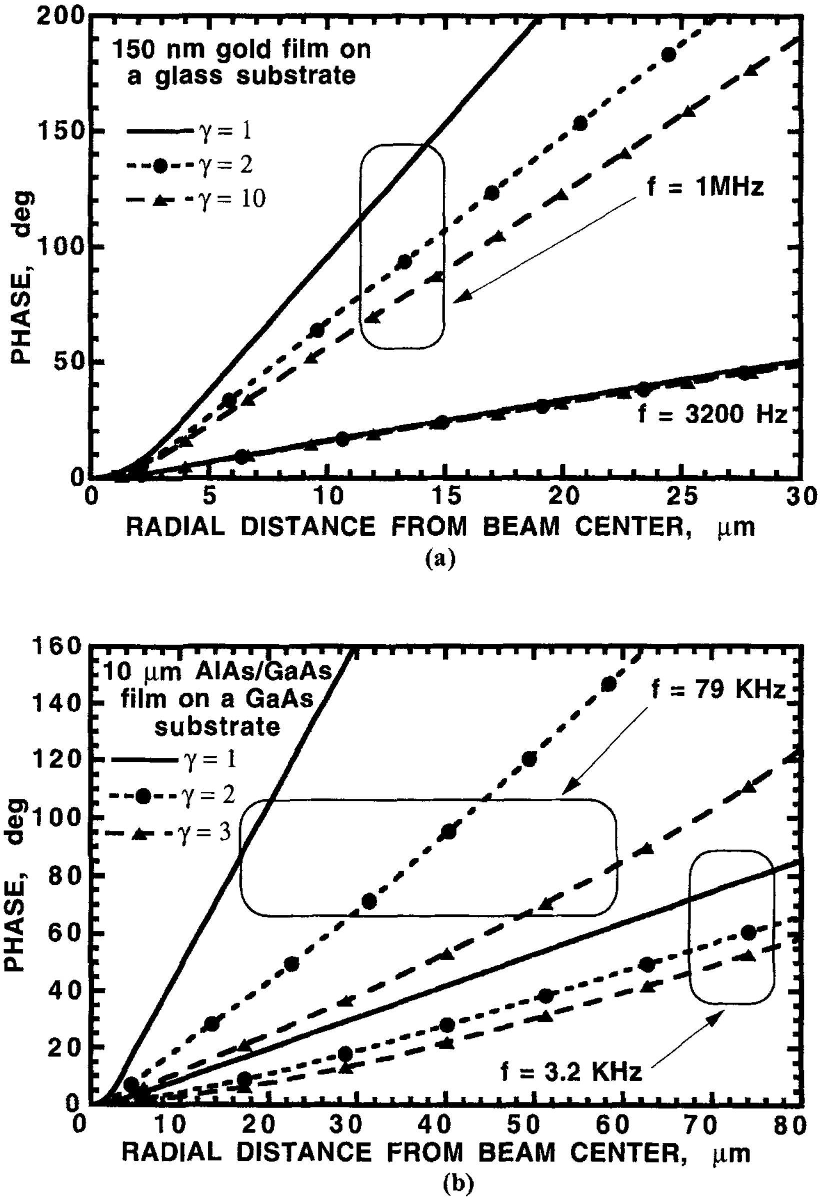
where α_s is the thermal diffusivity of the substrate and α_{fz} is the cross-plane thermal diffusivity of the film, k_s and k_{fz} are the corresponding thermal conductivities, ω is the angular modulation frequency, and γ is the ratio of the in-plane to the cross-plane thermal conductivity of the film.

Based on Eq. (1) it is possible to determine both the in-plane and the cross-plane thermal diffusivities, depending on which is the dominant heat conduction direction. The latter is influenced by several factors, including the film and substrate thermal conductivities, modulation frequency, laser beam radius, and location of detection. In the present experimental technique several of these factors can be controlled and thus used to facilitate the determination of the thermophysical properties. Figure 2a shows the theoretical phase signal variation as a function of the radial distance from the laser beam center, calculated for the case of a high-thermal conductivity film (a 150-nm film with an in-plane thermal diffusivity $\alpha_{||} = 0.96 \text{ cm}^2 \cdot \text{s}^{-1}$) on a low-thermal conductivity substrate (a glass substrate with a thermal diffusivity $\alpha = 3.4 \times 10^{-3} \text{ cm}^2 \cdot \text{s}^{-1}$), at low and high modulation frequencies. At low modulation frequencies the heat conduction inside the film is mainly in the in-plane direction and the anisotropy effect is negligible. The in-plane thermal conductivity can be determined from the radial distribution of the phase profile [7]. As the modulation frequency increases, the thermal wave is confined in a region close to the heat source and the temperature drop in the perpendicular direction is contained mainly in the film. Under this condition the anisotropy effect becomes important and the cross-plane thermal conductivity can be determined.

The case where the thermal conductivity of the film is lower than the thermal conductivity of the substrate is shown in Fig. 2b for a 10-μm-thick, 700 Å/700 Å GaAs/AlAs superlattice structure deposited onto a GaAs substrate. The in-plane film thermal diffusivity used in this calculation is $0.25 \text{ cm}^2 \cdot \text{s}^{-1}$ [12]. This value is relatively close to the value of the diffusivity for the film in Fig. 2a. However, the anisotropy determination for the superlattice film is much easier in the low-frequency range, since the thickness of the superlattice film is larger and comparable to the thermal diffusion lengths, $d \cong \sqrt{2\alpha/\omega}$. In general, the frequency range suitable for determining the anisotropic thermophysical properties for a film-on-substrate system depends on the film diffusivity, film thickness, and mismatch of thermophysical properties between film and substrate.

There is also another way to measure the cross-plane thermal diffusivity. If the laser beam diameter is large compared to the thermal diffusion length, and also the frequency range is such that the thermal diffusion length is comparable to the film thickness, the heat flow in the parallel direction is negligible compared to the heat flow perpendicular to the film. Determination of the cross-plane thermal conductivity is then possible from the frequency dependence of the thermal signal collected at the beam center.

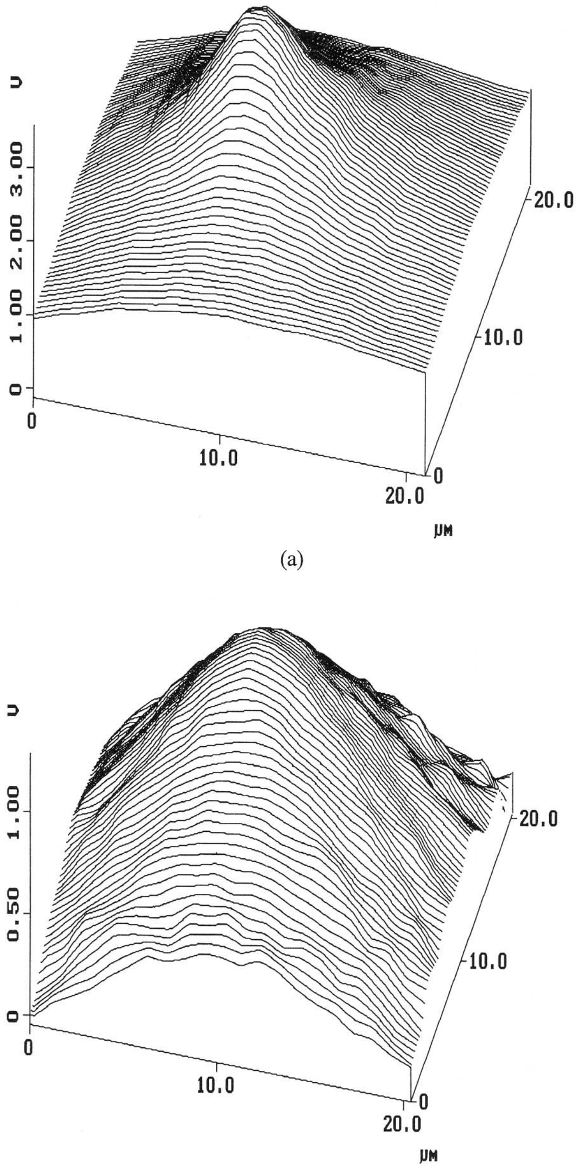
One other parameter that can affect the determination of the thermophysical properties for a film-on-substrate system is the interface thermal resistance. The effect of the interface thermal resistance on the determination of the film thermal diffusivity was ruled out for this experiment based on a simplified model developed by Hartmann et al. [7].



(a)

(b)

Fig. 2. Theoretically calculated phase signals at different modulation frequencies for (a) a 150-nm gold film on a glass substrate and (b) a 10-μm 700 Å/700 Å GaAs/AlAs superlattice on a GaAs substrate, where γ is the ratio of the in-plane to the cross-plane thermal conductivity for the film. The calculations assume a laser beam radius of 2 μm.



(a)

(b)

Fig. 3. Distribution of (a) the amplitude and (b) the phase of the thermal signal.

3. RESULTS AND DISCUSSION

Figure 3a shows the distribution of the amplitude and Fig. 3b the distribution of the phase of the experimental thermal signal collected at the surface of a 150-nm gold film evaporated on a glass substrate. The phase signal has a region of linear dependence on the laser beam displacement, in agreement with the recently reported photothermal reflectance detection method [7].

To obtain the thermal diffusivity of the film, the following fitting scheme is employed. First, the slope of the experimental phase signal in the linear region is determined. Then this slope is compared with the theoretically calculated slope of the phase signal using Eq. (1) for an estimate of the film thermal diffusivity. In these calculations, the thermophysical properties of window glass are taken for the substrate ($\alpha = 3.4 \times 10^{-3}\,\mathrm{cm}^2 \cdot \mathrm{s}^{-1}$, $k = 0.81\,\mathrm{W} \cdot \mathrm{m}^{-1} \cdot \mathrm{K}^{-1}$) [13]. For the given film and substrate thermophysical properties and at the given modulation frequency, the thermal diffusion length (about $100\,\mu\mathrm{m}$) is much larger than the laser beam diameter used in the experiment (about $4\,\mu\mathrm{m}$). Under this condition, numerical calculations show that the slope of the phase is independent of the laser beam size, as can be seen from Fig. 4. This behavior makes it possible to find the thermophysical properties of the film even if the laser beam diameter is not precisely known. With a film diffusivity of $0.96\,\mathrm{cm}^2 \cdot \mathrm{s}^{-1}$, the theoretical slope of the phase matches the experimentally

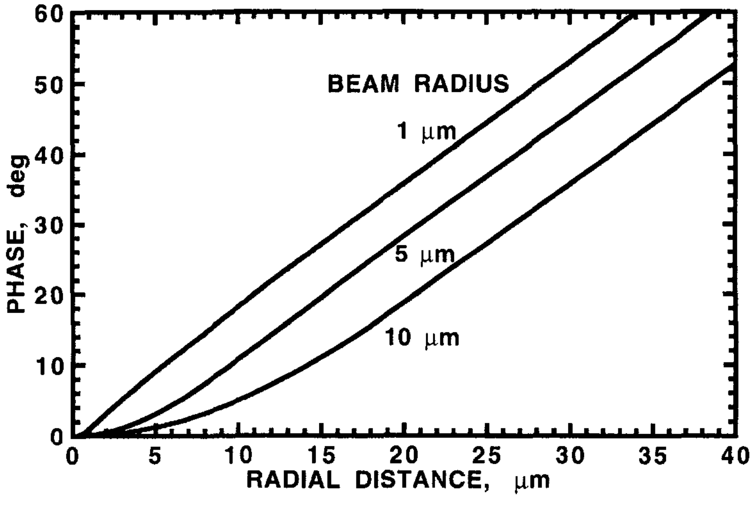


Fig. 4. Theoretically calculated phase signals showing that the slope in the linear region is independent of the laser beam radius.

determined slope. Figure 5a shows the phase fit and Fig. 5b the amplitude fit of the experimental signal for a thermal diffusivity of the film of 0.96 $cm^2 \cdot s^{-1}$. Hartmann et al. [7] reported a thermal diffusivity of 0.6 $cm^2 \cdot s^{-1}$ for a 160-nm sputtered gold film on quartz. Furthermore, the laser beam radius may be determined by fitting the experimental phase and

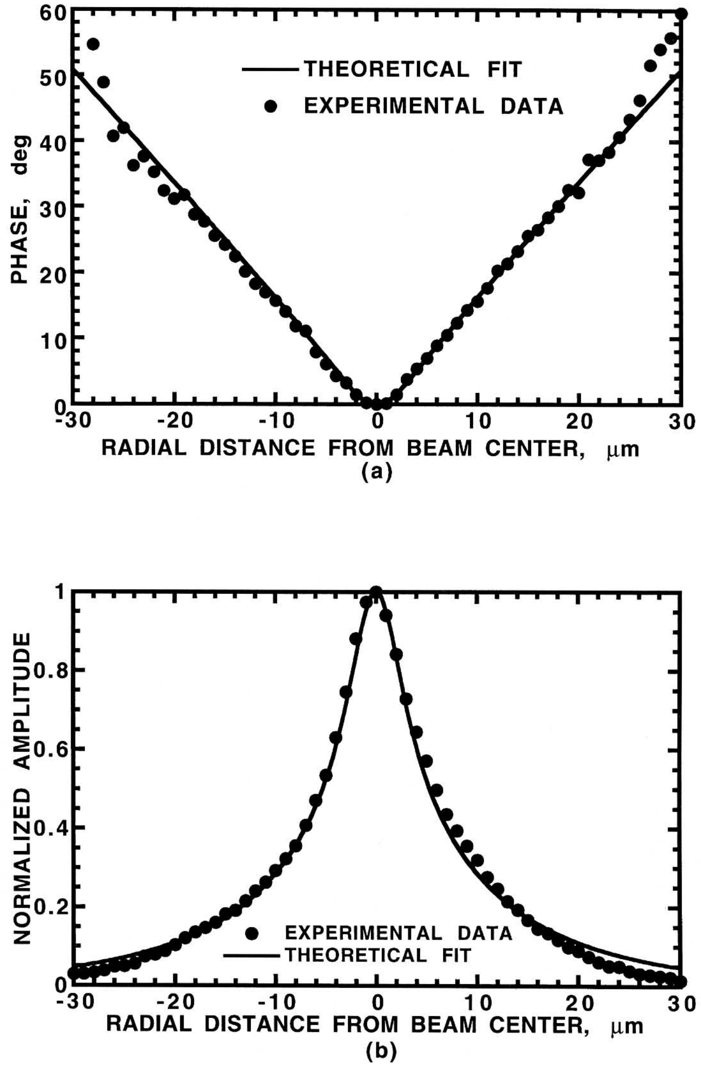


Fig. 5. Fitting of (a) the phase and (b) the amplitude of the experimental signal of a gold-on-glass sample with a film thermal diffusivity of 0.96 $cm^2 \cdot s^{-1}$.

normalized amplitude profiles with the theoretical calculations. The determined laser beam radius for this fit is 2 μm, which is close to the calculated 1-μm radius based on ABCD law [10]. For the current experiment, heat conduction inside the film is mainly in the in-plane direction and the determined thermal diffusivity is thus in this direction. As shown in Fig. 2a, higher modulation frequencies are required in order to detect any anisotropy effect for this particular sample.

The experiments were performed using a 3-ms time constant for the lock-in amplifier. In order to collect a less noisier signal, another run has been done using a 30-ms time constant. However, the thermal diffusivity value for the film derived from the data collected under this condition is $0.76\,\mathrm{cm}^2 \cdot \mathrm{s}^{-1}$. The discrepancy is attributed to the insufficient amount of time at each data point, i.e., about 39 ms, compared to the 30-ms time constant.

In the above experiments the gold film was deposited onto a glass substrate. However, in many situations the films are deposited onto semiconductor substrates and/or the films are nonmetallic. Many lasers are available to provide laser wavelengths at which semiconductor substrates are transparent [11]. For example, both Si and GaAs substrates are transparent at a 1.55-μm wavelength. High-power diode lasers at this wavelength are readily available. If the film is nonconductive, a very thin conductive film can be deposited onto the film of interest in order to make the temperature measurement possible. In the latter case, consideration must be given in the modeling and material selection to include the effect of heat conduction through the deposited film.

4. CONCLUSION

This work has demonstrated the potential of the scanning laser thermoelectric microscope for characterizing the thermophysical properties of thin films on a micrometer scale without removing the substrate. The microscope employs a modulated, focused laser as the heating source and a fast thermoelectric effect between the sample and a fine probe to detect the temperature response. Phase and amplitude images are presented for laser heating of a 150-nm gold film evaporated on a glass substrate. The thermal diffusivity of the film is obtained by fitting these signals with a three-dimensional heat conduction model.

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