Thermal conductivity of thin metallic films measured by photothermal profile analysis

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Thermal conductivity of nickel and gold films on quartz (thickness 0.4–8 μm) was measured by a modulated thermoreflectance technique recording the surface temperature profile. Model calculations predict an optimum frequency for measuring thermal transport within the film. Measurements on films with various thicknesses reveal a thermal conductivity close to the bulk value for nickel while gold films exhibit a reduced conductivity with decreasing film thickness. © *1997 American Institute of Physics.* [S0034-6748(97)04903-4]

I. INTRODUCTION

The growing interest in physical properties of thin films initiated the development of various techniques for a generally applicable, reliable, and noncontact determination of thin film thermal conductivity.[1] Most noncontact measurements are based on photothermal techniques utilizing modulated[2] or pulsed[3] laser beams for the determination of thermal conductivities. For a modulated laser beam as a heating source the solution of the heat diffusion equation yields thermal waves[4] with a one-dimensional decay length L_{th} depending on the thermal diffusivity D and the modulation frequency f:

$$L_{\text{th}} = \sqrt{\frac{D}{\pi f}}. \tag{1}$$

In the case of three-dimensional heat flow the decay length is smaller; i.e., the range is not only governed by the exponential decay $\exp(-r/L_{\text{th}})$ but also by an additional factor proportional to r^{-1}, where r is the distance from the heated center.[5]

Thermal wave propagation can be utilized for conductivity measurements by locally heating the surface with a harmonically modulated excitation and monitoring the surface temperature variation as a function of the distance from the heat source. Due to the propagation of the thermal wave the phase lag between the heat deposition and the temperature oscillation at a given point on the surface (or in the homogeneous material) is a linear function of the distance of the respective point from the heat source. Scanning over a thermal profile yields a straight line for the phase with a slope equal to the inverse of the thermal length.[6] This concept is the basis for thermal conductivity measurements presented here. However, since we investigate thin films on substrates and use a laser heating source with an extension that is not negligible compared to the width of the measured profile, data analysis is more complicated in our case and requires the use of a rigorous three-dimensional photothermal theory.[7]

From modulated heat flow experiments only the thermal diffusivity $D = \lambda/(\rho C_p)$ can be extracted. A conversion of thermal diffusivity D to thermal conductivity λ and vice versa is straightforward provided the specific heat ρC_p of the material is known. As the thermal conductivity is the more common quantity, in this work only thermal conductivities are presented, assuming that the heat capacity of the investigated thin films is equal to the respective bulk values.

II. EXPERIMENT

Imaging photothermal techniques are capable of directly measuring thermal decay in the surface plane[8] and, hence, allow a determination of thermal data by applying a theoretical model to the measured images. This is, e.g., exploited by methods suggested by Visser *et al.*[9] and Kemp *et al.*[10] who obtained two-dimensional thermal profiles induced by a local heat source from a modulated focused laser beam. In contrast to these techniques where the surface temperature distribution is measured by an infrared-scanner or a thermocouple, we scan the heating laser beam and measure the surface temperature distribution by thermoreflectance[11] with an apparatus originally designed for defect mapping in optical thin film systems.[12] The experimental arrangement is schematically depicted in Fig. 1. The modulated heat source is a mechanically chopped beam from an Ar^+ laser operated at 514 nm that can be positioned with μm resolution by means of two dielectric mirrors mounted on computer-controlled translation stages. To obtain an undistorted Gaussian beam profile with a width of 100 μm at the sample surface, the laser beam is spatially filtered by a pinhole in the focal plane of a Galilean telescope. Since thin films are easily damaged by excessive heating it is essential to monitor the incident power, accomplished in our case by a beam splitter directing part of the laser beam to a calibrated sensor head. The thermoreflectance is probed by a HeNe laser beam focused to about 30 μm at the same location as the heating beam. This ensures that convolution effects resulting from the finite width of the pump laser beam and the 45°-incidence of the probe beam can be neglected. After narrow-band spectral filtering the reflected probe beam intensity is measured by a photodiode coupled to a current-to-voltage converting preamplifier. The harmonic component of the modulated reflectivity signal is detected by a two-phase lock-in amplifier that directly provides amplitude and phase of the sample surface temperature oscillation. By means of measuring a reference

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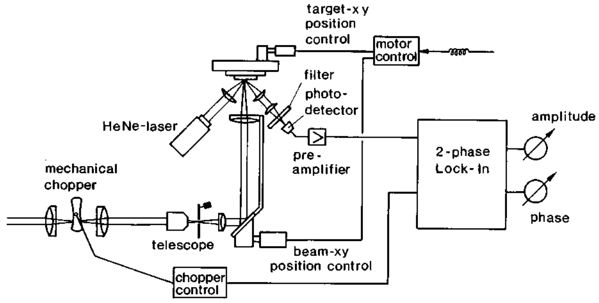

FIG. 1. Schematic view of the experimental setup used for thermoreflectance measurements.

material the amplitude can easily be calibrated absolutely; however, this was not necessary for the measurements presented here. With this arrangement thermal profiles are taken by moving the heating beam with respect to the probing beam that remains positioned at a fixed location of the sample surface. This requires thermally homogeneous sample properties over the scanned region. To select a suitable surface region, the sample can also be moved parallel to the surface plane by translation stages.

III. RESULTS

As the frequency dependent thermal length is a crucial parameter for the thin film investigation it must be carefully chosen for a specific sample thickness. To find a region of optimum film sensitivity we carried out model calculations of the width of the surface temperature profile for various modulation frequencies using the theory outlined in Ref. 7. Figure 2 shows results for a nickel layer on a quartz substrate for modulation frequencies of 100 Hz and 1 kHz, respectively. The full width of the amplitude profile at 1/8 of the peak temperature is plotted as a function of film thickness. Vertical lines indicate a film thickness equal to the thermal length L_{th}.

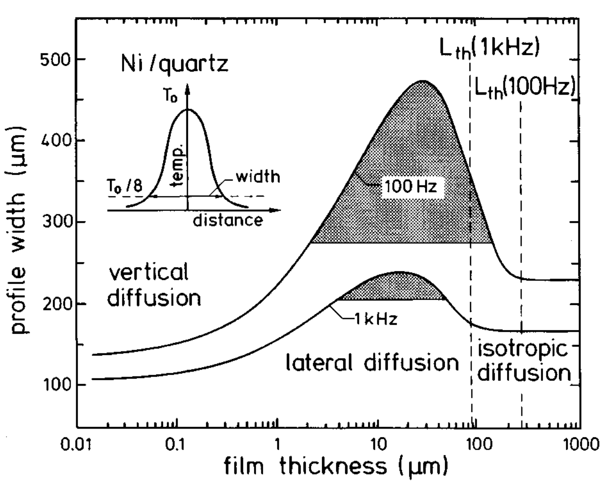

FIG. 2. Model calculations for the width of the photothermal amplitude profile at 1/8 of the maximum for nickel films on quartz. Vertical lines indicate a thickness equal to the thermal length at 100 Hz and 1 kHz, respectively. The shaded areas denote regions of optimum film sensitivity. These regions are defined by a thermal broadening of at least 20% with respect to the isotropic diffusion limit.

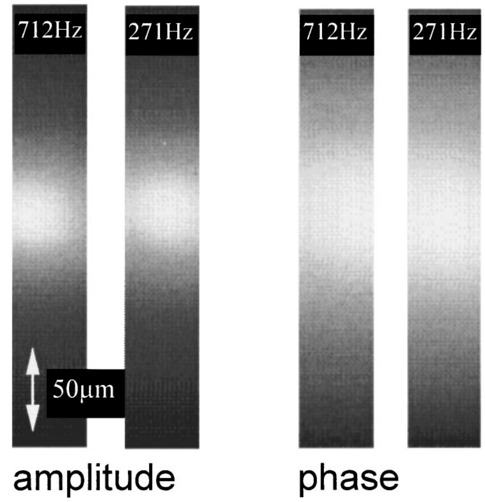

FIG. 3. Two-dimensional profiles of amplitude and phase of the photothermal signal for a 1.69 μm gold film on quartz at 271 and 712 Hz modulation, respectively. Gray scaling from black to white represents zero to maximum for the amplitude and a shift of 60° for the phase.

For a film thickness large compared to L_{th}, the photothermal response is similar to that of a bulk material and we find isotropic diffusion without any thickness dependence. For a reduced film thickness, the heat preferentially expands laterally as soon as the thermal diffusion length exceeds the film thickness since the heat flow into the depth of the sample is affected by the poorly conducting quartz substrate. The sensitivity of the measurement to heat flow within the film is increased as the width of the temperature profile rises until an optimum film sensitivity is achieved at about 20–30 μm film thickness for the assumed frequencies. In the vicinity of the optimum frequency the heat flow can be well described by a simplified model allowing a straightforward determination of thermal parameters.[13] This optimum range (indicated by the shaded regions in Fig. 1) narrows and shifts towards smaller thickness for higher frequencies. For a thickness below the optimum range the film is no longer able to carry away the heat and the heat flow is dominated by vertical diffusion into the substrate. The width of the profile for the asymptotic cases of very thick and thin films represent the values for film and substrate bulk material, respectively.

Principally, the diffusivity could be extracted from amplitude profiles as calculated above. However, as the phase of the modulated signal is generally less sensitive to experimental errors than the amplitude in our work the former was used for diffusivity determination presented in this work. For our investigations we used gold and nickel films of 0.4–8 μm thickness sputtered on quartz substrates. This range was chosen since we expected thickness dependent changes in thermal conductivity below about 1 μm. However, film preparation was restricted to a thickness below 2 μm for gold films since internal stress in these films with a very weak adhesion to the substrate caused delamination for thicker films. From Fig. 2 it is evident that with a setup operated at frequencies up to 1 kHz it is not possible to work in the optimum film sensitivity region; however, the use of the complete three-dimensional theory assured reliable data analysis.

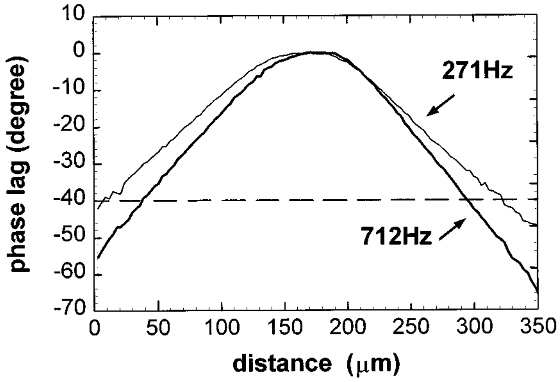

FIG. 4. Cross sections through the phase profiles from Fig. 3. The dashed line indicates the 40° phase change used to define the width of the profiles.

As an example, data analysis for a gold-film of 1.69 μm thickness is described in some detail. Two-dimensional thermoreflectance profiles have been taken by rastering the pump beam while the probe beam remained at a fixed position. Figure 3 shows amplitude and phase profiles at 271 and 712 Hz, respectively. Cross sections of the two-dimensional phase data array were taken, yielding profiles through the maxima of the phase curves as shown in Fig. 4. The profiles exhibit a linear decrease at some distance from the center while the behavior in the center region is determined by the 100 μm width of the Gaussian heating beam. The full widths of the phase curves at 40° change (indicated in Fig. 4) by a dashed line were extracted as a measure for thermal broadening. The frequency dependence of this broadening is displayed in Fig. 5. For comparison, three theoretical values for each measured width are shown, namely the bulk literature value of 318 W/mK and this value plus or minus 10%. From this picture a thermal conductivity of 314 W/mK is deduced. The overall error for the obtained conductivity values is estimated to be less than 20%, corresponding to the reproducibility of the results and possible systematic errors, e.g., introduced by neglecting a thermal resistance at the interface between thin film and substrate.

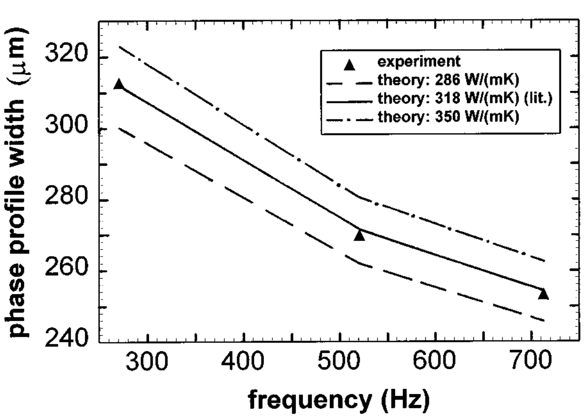

FIG. 5. Frequency dependence of the measured phase widths at 40° for the 1.69-μm-thick gold film on quartz. The dashed lines indicate theoretical predictions for 286, 318, (literature value), and 350 W/mK.

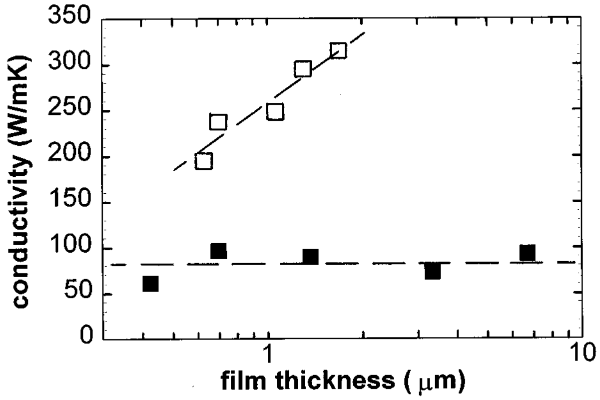

FIG. 6. Dependence of thermal conductivity on layer thickness for gold (open squares) and nickel films (filled squares) on quartz. Dashed lines are a guide to the eye.

IV. DISCUSSION

Results for a series of gold and nickel films obtained by this method are shown in Fig. 6, where the conductivity has been plotted as a function of film thickness. It can be seen that thermal conductivity decreases with decreasing film thickness in the case of gold films. This observation can be explained by the polycrystalline structure of sputtered films,[14] where thermal barriers between the grains affect the lateral heat flow resulting in a lower thermal conductivity. Similar effects have also been observed with other noncontacting[15] and contacting conductivity measurements.[1,16–18] Although the grains grow with increasing film thickness, conductivities of the thickest investigated films are still slightly lower than bulk values because the polycrystalline structure affects heat flow even in the thickest films. Apparently, thermal transport in nickel films is much less affected by these effects since a conductivity close to the bulk value of 91 W/mK is found throughout the thickness range investigated.

To evaluate the feasibility of the experimental technique presented here for the measuring of thin film thermal conductivities it has to be compared to other experimental approaches presented in the literature. A major feature of the thermoreflectance technique is its ability to measure thermal properties with high spatial resolution, which is a great advantage when working with metallic microstructures; it may, however, also be a disadvantage when averaged information about an extended film is sought. The only alternative technique likely to obtain a similar resolution is the thermal comparator method[18] that often suffers from great uncertainty and systematic errors due to a poor mechanical contact between the comparator and the sample. The thermoreflectance method can be applied *in situ* to samples of arbitrary geometry and does not change or destroy the sample. This is in contrast to thin film thermal conductivity measurements based on thermometry,[16] where heaters and sensors have to be attached to or evaporated onto the samples. A simple contacting method for the determination of thermal conductivity in the plane of the film is a measurement of its resistance and conversion of this data to thermal conductivity exploiting the Wiedemann–Franz relationship.[19,20] In such measurements, however, only the electronic part of the thermal conductivity is measured and the lattice contribution has to be estimated independently.[20] It has been pointed out[19] that the phonon contribution to conductivity may be high for thin films compared to bulk metals due to lattice imperfections. Therefore, appropriate correction factors are difficult to determine for thin films and may introduce considerable errors.

V. CONCLUSION

In conclusion it was shown that the modulated temperature profile analysis is a versatile tool to study thermal conductivity of highly conducting thin films on a poorly conducting substrate. An optimum modulation frequency regime for thermal profile measurements was defined. Using a complete three-dimensional theory for data analysis yields reliable results also outside this region, provided the substrate parameters are well known. Measurements on films with various thickness revealed a thickness dependence of thermal conductivity in the case of gold films on quartz substrates, while nickel film conductivity was found to be independent of thickness.

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