Picosecond Ultrasonic and Heat Flow Measurements with Enhanced Sensitivity

T. J. Grimsley,[1] J. Ma,[1] Q. Miao,[1] A. V. Nurmikko,[1,2] and H. J. Maris[1,*]

[1]*Department of Physics, Brown University, Providence, Rhode Island 02912, USA*

[2]*Division of Engineering, Brown University, Providence, Rhode Island 02912, USA*

(Received April 11, 2010)

We report on two variations of the standard measurement technique used for making picosecond ultrasonic and heat flow measurements. In the first variation, a resonant optical cavity is produced above the surface of the sample. For a highly reflecting sample, this cavity enhances the fraction of the energy of the pump pulse that is absorbed by the sample, and also increases the change in the reflection of the probe pulse that results from the change in the optical constants of the sample. In the second variation, a transparent optical mask is placed just above the surface. This mask results in a periodic variation of the intensity of the pump and probe beams across the sample surface, and makes possible the study of lateral heat flow in thin films and nanostructures.

PACS numbers: 78.47.D-, 42.62.Eh, 43.35.Ns

I. INTRODUCTION

In this paper we report on two experiments in which modified versions of the conventional pump and probe technique for ultrasonic and thermal measurements are used.

II. EXPERIMENTS WITH AN OPTICAL CAVITY

In a conventional picosecond ultrasonic experiment, a sound pulse is generated when a light pulse ("pump pulse") is absorbed in a sample [1]. When an acoustic echo returns to the surface of the sample, there is a small change $\Delta R(t)$ in the optical reflectivity of the sample; this change is measured with a time-delayed probe light pulse. The magnitude of $\Delta R(t)$ is typically on the order of 10^{-5}, but in some situations it may be smaller. To measure $\Delta R(t)$, it is necessary to use lock-in detection and signal averaging to achieve reasonable signal to noise.

The change in the reflected intensity comes about because the strain of the acoustic pulse results in a change in the optical constants of the material near to the sample surface; the magnitude of this change is determined by the piezo-optic coefficients of the material. For this reason, one cannot carry out picosecond ultrasonic experiments when these coefficients are very small. For example, many commercial ultrafast lasers operate at

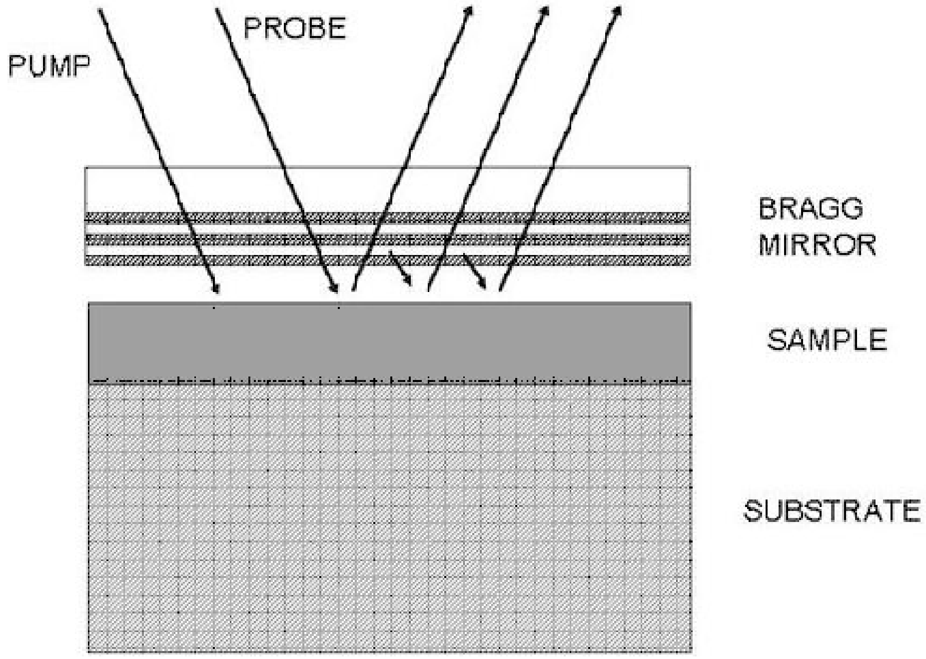

FIG. 1: Schematic diagram of the optical cavity apparatus.

a wavelength near to 800 nm, a wavelength at which wavelength the piezo-optic coefficient for copper is nearly zero [2]. To make measurements in this situation, one possibility is to determine the change in the phase of the reflected probe light, rather than the change in the intensity. The phase change arises from the displacement of the sample surface. Several different types of interferometer have been used [3–9]. Normally, the fractional change in the output signal from the interferometer is of the same order of magnitude as the change in the phase, i.e., again one is making a measurement of a change in signal of the order of 10^{-5} or less.

In this paper, we report on experiments in which we place a mirror (distributed Bragg reflector DBR) above the sample surface so as to form a resonant optical cavity. The characteristics of the mirror can be chosen so as to enhance the absorption of the pump light pulse and thereby to increase the amplitude of the generated sound. The cavity can also be designed so as to give a large increase in the reflectivity change $\Delta R(t)$. The effectiveness of this technique can be improved by using slightly different wavelengths for the pump and probe light, or different angles of incidence as described below. This make it possible to maximize the absorption of the pump light while making the reflectivity of the probe change by a large amount when the cavity spacing is changed by the displacement of the surface of the sample.

The experiment is shown schematically in Fig. 1. The DBR has a reflectivity of 0.84 for 800 nm light, and the sample is a copper film deposited onto a silicon wafer. The DBR was mounted on a precision stage. This stage made it possible to maintain the separation

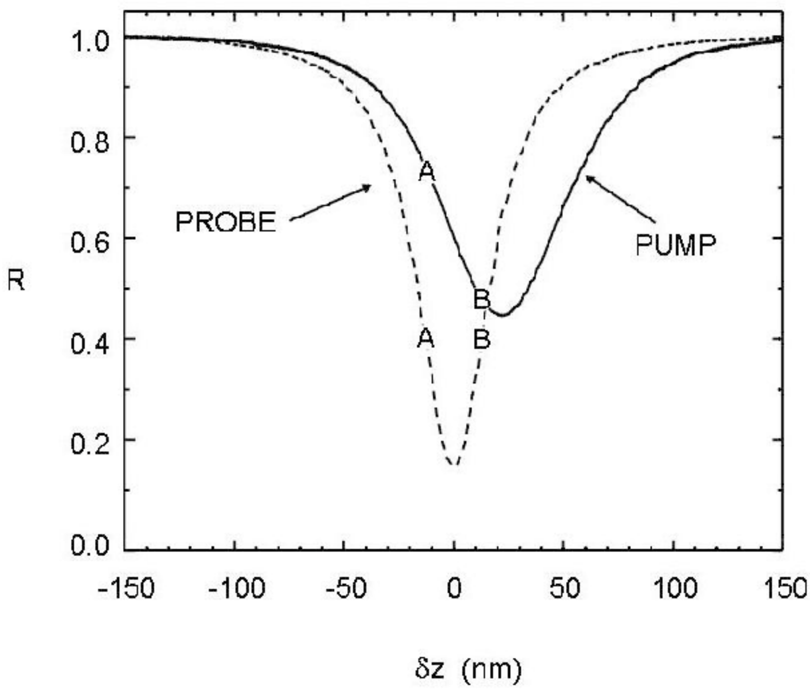

FIG. 2: Optical reflectivity of the pump (dashed line) and probe (solid line) as a function of the displacement of the dielectric Bragg reflector from a reference position. Measurements were made for cavity spacings A and B.

between the DBR and the sample at an accuracy of approximately ±1 nm; the angular orientation of the DBR was parallel to about 100 microradians. The pump and probe beams were focused onto spots of diameter approximately 20μ, and so with the angular control, the variation in the cavity spacing across the area of these spots was on the order of 2 nm.

In Fig. 2, we show the results of measurements of the reflectivity of the pump and probe light in the vicinity of a cavity resonance as a function of a change in the cavity spacing from a reference position. For these data, the probe light was at normal incidence, and the pump was at an angle of 8° from normal. The probe light was passed through a line filter of bandwidth 3 nm to reduce the spread of wavelengths and give a sharper resonance. By having the pump and probe at different angles, we can find a cavity spacing such that the absorption of the pump is large, and the reflection of the probe changes rapidly with a change in the cavity spacing.

For a DBR, the phase of the reflected light is not equal to the phase that would result from reflection at the actual physical surface of the DBR. Thus, knowledge of the distance between the surface of the DBR and the surface of the sample cannot be used to make a definite determination of the order of a particular cavity resonance. We estimate that for the resonance shown in Fig. 2, the physical distance between the DBR and the surface of the

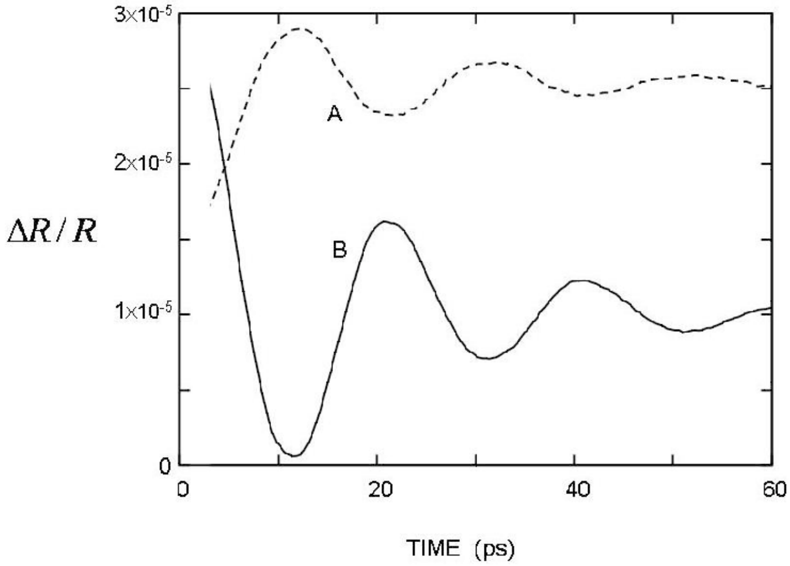

FIG. 3: Reflectivity change $\Delta R(t)/R$ as a function of the time delay of the probe light pulse. These data were taken for cavity spacings A and B as indicated in Fig. 2.

sample is about 2μ, and that the resonance is the 4th or 5th cavity mode. The sharpness of the resonance is limited by the finite reflectivity of the DBR and the sample, and by the spectral width of the laser pulse. The resonance is further broadened because of the range of propagation angles of the light making up each beam; this has a much larger effect for the pump than for the probe because the pump is not at normal incidence. The effect of the spectral width and the angular spread increases as the order of the cavity resonance increases, and so it is advantageous to work with as low a cavity mode as possible. But in order to do this, it is necessary to have the surfaces of the sample and the DBR free of any contaminant particles.

In Fig. 3, we show data obtained with a sputtered copper film with a thickness of 47 nm deposited onto a silicon substrate. The data were taken for the cavity spacings indicated by A and B in Fig. 2. One can see from Fig. 2 that at these two cavity spacings, the rate of change of the probe reflectivity has the opposite sign but approximately the same magnitude. Signal B is larger because the absorption of the pump is larger, as can be seen from Fig. 2.

In a recent paper [10], we have given a detailed theoretical analysis of the effect on the sensitivity of varying the reflectivity of the DBR, and have analyzed how the sensitivity is affected by the finite bandwidth of the pump and probe pulses.

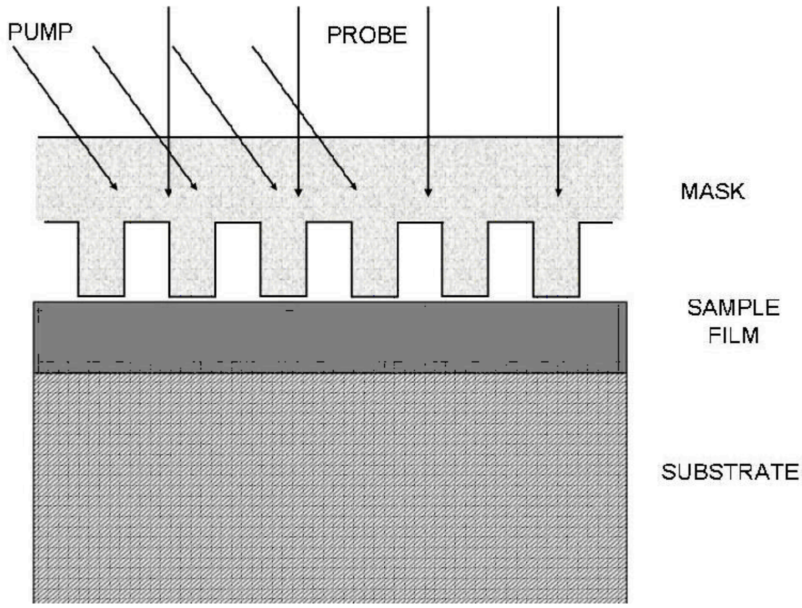

FIG. 4: Schematic diagram of the apparatus for studying lateral heat flow.

III. EXPERIMENT WITH AN OPTICAL MASK

In a conventional picosecond ultrasonic experiment, the pump and probe beams are each focused onto a spot on the sample surface that has linear dimensions of about 10μ. Since the spot is usually large compared to the thickness of the sample, it is adequate to treat the sound propagation and heat flow as one-dimensional problems, although there are some cases where this is inadequate. In soliton experiments, for example, the propagation velocity is dependent on the strain amplitude and so the sound generated at different regions within the pump spot travels at different speeds. There would be many advantages of extending the basic pump and probe method so that one could study the propagation of sound or heat in the lateral direction. There are a number of possible ways in which this might be achieved. The spot size for the pump beam could be reduced [11, 12], thereby generating a surface wave spreading outwards across the surface; this could be detected by a probe beam focused at some distance from the pump. One could modify the sample so that even though the pump and probe beams are not sharply focused, the pump and/or the probe light would be absorbed to a different extent in different areas of the sample surface [13–15]. The disadvantage of this is that the sample has to be modified. Finally, one can use the transient grating technique in which the pump beam is divided into two components that are incident on the sample at different angles and produce an interference pattern across the surface [16]. If these beams are at angles of $\pm\theta$ from normal incidence, the intensity has a period of

$$\tilde{\lambda} = \frac{\lambda}{2\sin\theta}.$$

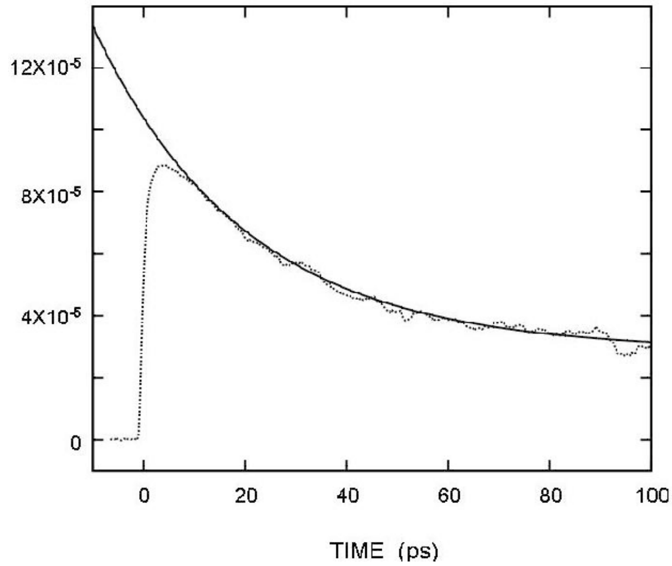

FIG. 5: Experimental data obtained with the optical mask (dotted curve). The solid line shows the fit to the data as described in the text.

Thus, for 800 nm light and $\theta = \pi/4$, the period is 560 nm.

To achieve a shorter period than can easily be achieved through interference of two light beams, we have used the setup shown schematically in Fig. 4. A transparent mask is placed above the sample and the pump, and probe beams pass through this mask. Because of the pattern of the mask, the intensity of the light reaching the sample varies periodically across the sample surface. The SiO_2 mask that was used for the measurements that we report here consisted of a series of lines with a period of 200 nm, and the depth of the trenches between the lines was 200 nm. The light wavelength was 800 nm, with the pump at 45° from normal incidence and polarized with the electric field parallel to the lines, and the probe at normal incidence and polarized perpendicular to the lines. It can be expected that as the distance of the mask from the surface of the sample increases, the amplitude of the variations of the intensity with position on the sample surface will rapidly decrease. Thus, in order to provide spatially patterned heating of the sample, it is essential to have the mask at a distance from the sample that is less than the repeat distance of the mask. In previous work in our laboratory, measurements were made with a mask of period 1μ [14]. For the 200 nm mask used here, getting the mask sufficiently close to the sample surface is a significant experimental challenge. We will describe work on this in a separate paper.

n Fig. 5, we show data taken with the mask on an Al film of thickness $d_{Al} = 20$ nm deposited onto a GaAs substrate. The measured quantity is the change ΔR in the optical reflectivity as a function of the time delay t of the probe relative to the pump. The data in the time range out to 100 ps can be well fit by the expression

$$\Delta R(t) = 2.9 \times 10^{-5} + 7.5 \times 10^{-5} \exp(-t/30). \quad (1)$$

We can understand this as follows. If measurements were made without the mask so that the Al film was heated uniformly, there would be a change in optical reflectivity proportional to the temperature change $\Delta T(t)$ (thermoreflectance). The film would cool by conductance across the interface into the GaAs. The rate of cooling would be governed by the Kapitza conductance σ_K, and so

$$\Delta T(t) = \Delta T(t=0)\ \exp(-t/\tau_K), \tag{2}$$

where

$$\tau_K = C_{Al} d_{Al}/\sigma_K, \tag{3}$$

and C_{Al} is the specific heat per unit volume of Al. A reasonable value to expect for σ_K is 10,000 W $\text{cm}^{-1}\text{K}^{-1}$, and this leads to the value 480 ps for τ_K. In fact, the cooling will be slower than this because of the thermal resistance of GaAs. Thus cooling by heat flow into the substrate does not explain the component on $\Delta R(t)$ that decays with a time constant of 30 ps, but only contributes to the decrease in $\Delta R(t)$ at much longer times (approximately represented by the constant term in Eq. 3). It appears that the 30 ps component originates from the lateral heat flow in the Al. To analyze this we first note that for a 20 nm Al film, the heat deposited by the pump pulse will spread throughout the thickness of the film in a time which is of the order of 1 ps. The time for a lateral temperature variation with periodicity L to relax is governed by the time constant

$$\tau_{lateral} = \frac{C_{Al} L^2}{4\pi^2 \kappa_{film}} \tag{4}$$

where κ_{film} is the thermal conductivity of the film. Using $L = 200$ nm, the value required to give the measured value of 30 ps is $\kappa_{film} = 0.82$ W $\text{cm}^{-1}\text{K}^{-1}$. This compares with the standard value for the thermal conductivity of bulk Al of $\kappa_{Al} = 2.37$ W $\text{cm}^{-1}\text{K}^{-1}$. A significant reduction in the thermal conductivity for a film relative to the conductivity for bulk material is to be expected. We had hoped to check the experimental result through the measurement of the electrical conductivity σ_{film} followed by the use of the Weidemann-Franz law to estimate κ_{film}, but it is extremely difficult to make a reliable measurement of σ_{film} for a thin and very soft film of aluminum.

It is worthwhile to note that the use of a mask with a short repeat distance is essential for this measurement of the thermal conductivity. If we had used a mask with repeat distance 800 nm, or the interference of two pump beams to make a grating with this period, the time $\tau_{lateral}$ would have increased to 480 ps. Then the rate of decay of $\Delta R(t)$ due to lateral heat flow and heat flow into the substrate would be comparable, and it would be very difficult to extract the thermal conductivity of the film.

At present, we have no quantitative theory of the magnitude of the signal ΔR when an optical mask is used. To construct such a theory, it is necessary to consider several different physical effects. The change in the optical reflectivity of the Al film gives one contribution. However, it is also necessary to include the effect of the surface displacement of the Al which changes the distance between the film surface and the mask (the initial value of this separation is not reliably known). A temperature-induced change in the phase of the optical reflectivity coefficient of the film will have an effect similar to that of surface displacement and also needs to be allowed for.

IV. SUMMARY

We have reported on measurements made using two variations of the basic picosecond ultrasonic and heat flow technique. These variations make possible ultrasonic measurements on materials that show no significant piezo-optic response, and the study of lateral heat flow in thin metallic films.

Acknowledgments

This work was supported in part by the Air Force Office of Scientific Research under Contract No. FA9550-08-1-0340 and by Rudolph Technologies, Inc.

References

* Electronic address: humphrey_maris@brown.edu
[1] C. Thomsen, H. T. Grahn, H. J. Maris and J. Tauc, Phys. Rev. B **34**, 4129 (1986).
[2] U. Gerhardt, Phys. Rev. **172**, 651 (1968).
[3] D. H. Hurley and O. B. Wright, Opt. Lett. **24**, 1305 (1999).
[4] C. J. K. Richardson, M. J. Ehrlich, and J. W. Wagner, J. Opt. Soc. Am. B **16**, 1007 (1999).
[5] M. Nikoonahad, S. Lee, and H. M. Wang, Appl. Phys. Lett. **76**, 514 (2000).
[6] C. Glorieux, J. D. Beers, E. H. Bentefour, K. Van der Rostyne, K. A. Nelson, Rev. Sci. Inst. **75**, 2906 (2004).
[7] C. Rossignol and B. Perrin, IEEE Transactions on Ultrasonics, Ferroelectrics and Frequency Control **52**, 1354 (2005).
[8] T. Tachizaki, T. Muroya, O. Matsuda, Y. Sugawara, D. H. Hurley and O. B. Wright, Rev. Sci. Inst. **77**, 043713 (2006).
[9] S. Bramhavar, B. Pouet, and T. W. Murray, Appl. Phys. Lett. **94**, 114102 (2009).
[10] Y. Li, Q. Miao, A. V. Nurmikko, and H. J. Maris, J. Appl. Phys. **105**, 083516 (2009).
[11] C. Rossignol, J. M. Rampnoux, M. Perton, B. Audoin and S. Dilhaire, Phys. Rev. Lett. **94**, 166016 (2005).
[12] T. Dehoux, N. Chigarev, C. Rossignol, and B. Audoin, Eur. Phys. J. Special Topics **153**, 231 (2008) and Phys. Rev. B **77**,214307 (2008).
[13] D. H. Hurley and K. L. Telschow, Phys. Rev. B **66**, 153301 (2002).
[14] G. A. Antonelli, P. Zannitto, and H. J. Maris, Physica B **316**, 377 (2002).
[15] B. C. Daly, N. C. R. Holme, T. Buma, C. Branciard, T. B. Norris, D. M. Tennant, J. A. Taylor, J. E. Bower, and S. Pau, Appl. Phys. Lett. **84**, 5180 (2004).
[16] See, for example, R. M. Slayton and K. A. Nelson, J. Chem. Phys. **120**, 3908 (2004).