

STUDIES OF VERY SMALL STRUCTURES USING PICOSECOND ULTRASONICS

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Humphrey_Maris@brown.edu**Abstract**

Picosecond ultrasonics is a technique that makes ultrasonic measurements at high frequency (up to around 1000 GHz) possible with excellent time resolution. We give a short review of the development of this technique, beginning with a description of the mechanism for the generation and detection of sound, and then discussing some of the experiments that have been performed. Finally, we mention some of the practical applications of picosecond ultrasonics.

Introduction

In any pulse-echo ultrasonic experiment, it is essential to use sound pulses with a spatial length that is less than the round trip distance through the sample. This is to avoid the overlap of successive echoes. As a consequence, in order to make measurements on very small structures it is necessary to generate and detect very short acoustic pulses. In many ultrasonic measurements sound is generated by means of a piezoelectric transducer that is driven by an rf pulse. The length of this pulse is often several μs , but may in special circumstances be as short as a few ns. Even for an rf pulse of duration 3 ns, the spatial length of the sound pulse generated in a material with sound velocity $5 \times 10^3 \text{ m s}^{-1}$ is at least $15 \mu\text{m}$. Thus, this type of experimental approach is not suited for measurements on samples whose thickness is around $10 \mu\text{m}$ or less. In this article we describe a method, picosecond ultrasonics [1,2,3], that enables ultrasonic measurements to be made on structures in which the round trip distance is as short as 10 nm.

Generation and Detection of Sound

In this method short light pulses are used to generate and detect sound (Fig. 1). The sample is a thin film that has been deposited onto a substrate. When a "pump" light pulse is focused onto an area of the sample surface, the absorption of the light raises the temperature of a layer near the surface by a few degrees. This sets up a thermal stress distribution and a sound pulse propagates away from the surface. This sound pulse will be partially reflected at the interface between the sample film and the substrate. When the echo returns to the top surface of the sample film, it is detected by means of a probe light pulse.

Before considering the experiments that can be performed using this method, we explain the generation and detection mechanism in more detail. Pulsed lasers can now provide light pulses as short as 6 fs, this time corresponding to just a few cycles of oscillation of the light wave. On this time scale, it is a substantial oversimplification to believe that the light pulse "heats" the surface of the sample [4]. If the sample is a metallic film, the energy in the light pulse is first given to the conduction electrons, raising some number of them to states with energy above the Fermi energy E_F and creating an equal number of holes below E_F . This change in the energy distribution of the conduction electrons sets up a stress (electronic stress) in the sample which is proportional to the deformation potential. The electrons then begin to lose energy to the phonon system, and it is only after this process is completed that the system is described by a temperature. At the same time that the transfer of energy from the electrons to the phonons is taking place, the hot electrons are diffusing rapidly into the interior of the sample. As a result, even though the light is absorbed over a very short distance from the sample surface (e.g., 10 nm), by the time the energy reaches the phonon system the energy may have penetrated a much greater distance into the sample (e.g., 100 nm). The shape of the generated sound pulse is determined by the temporal and spatial dependence of the transient stress set up in the sample by the light pulse. Consequently, both the time for energy transfer between the electrons and the phonons and the rate of electron diffusion have an important effect on the shape of the generated sound pulse.

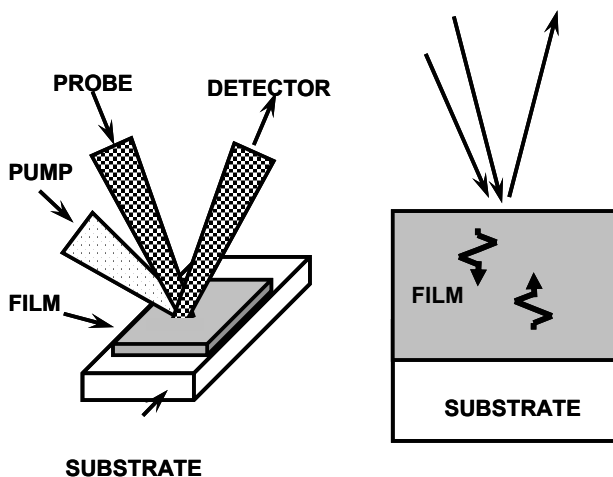


Figure 1. Schematic diagram of a picosecond ultrasonic experiment.

In most experiments, the diameter of the region illuminated by the pump light pulse is large

compared to the spatial length of the sound pulse that is generated and the intensity of the pump light varies slowly over this region. As a result, a longitudinal acoustic pulse is produced that propagates into the sample without appreciable diffraction. By modifying the experimental setup, the intensity of the pump light can be made to vary periodically across the sample surface. The stress resulting from the pump pulse then leads to the generation of surface waves in addition to the bulk waves that propagate into the sample. A periodic variation of the intensity of the pump light can be produced in several different ways. In one approach, the pump light pulse is split into two components that are directed at the surface from different angles so that they form a periodic interference pattern on the sample surface [5]. Alternatively, the pump light can be directed through an optical mask that consists of a series of narrowly spaced lines [6]. If this mask is positioned just above the surface of the sample, the intensity of the pump light beam will vary periodically across the sample surface and surface waves will be generated.

The details of the detection mechanism are also interesting. In most picosecond ultrasonic experiments performed so far, the arrival of the returning sound echo has been detected through a measurement of the change in the intensity of the reflected probe light. This change arises because the optical “constants” of the material n and κ are modified by the elastic strain of the sound pulse. However, it is important to realize that the normal component of the stress at a free surface is always zero. Since we are dealing with a longitudinal sound pulse propagating at normal incidence to the surface, it follows that all components of the elastic strain tensor at the surface are also zero, and there is no change in n and κ at the surface. The change in the reflected intensity of the probe comes about because the light penetrates a small distance into the sample and so reaches a region where n and κ are changed. The theory of this is given in ref. 1.

The variation of n and κ with strain is determined by the piezo-optic coefficients (POC). Measurements of these POC parameters for a number of materials are presented in ref. 7. For many materials, the POC have a complicated and sometimes rapid variation with wavelength. This has the consequence that for a given wavelength of the probe light pulse, much larger signals are detected in some materials than in others [8,9]. For example, with 800 nm light echoes are easy to detect in Al, but there is almost no signal from Cu. However, it is straightforward to use a frequency doubler to convert the laser output to 400 nm, and for this wavelength a strong signal is obtained from Cu.

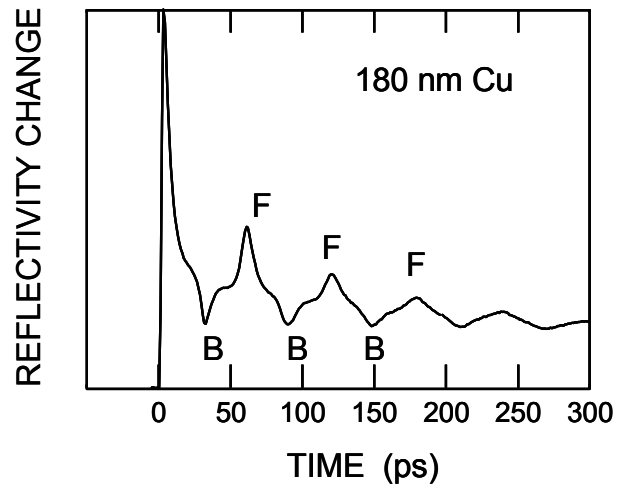


Figure 2. Results of an ultrasonic measurement in a 180 nm film of copper. The echoes labeled F and B arise from strain pulses generated at the front and back of the sample, respectively.

The change ΔR in the optical reflectivity can be written as [1]

$$\Delta R(t) = \int f(z) \eta(z,t) dz, \quad (1)$$

where $\eta(z,t)$ is the strain in the sample at a distance z below the surface at time t , and $f(z)$ is a function that depends on the POC. If the POC are known, it is possible to use the measured $\Delta R(t)$ together with Eq. 1 to estimate the shape of the sound pulse.

The acoustic echoes can also be detected by measuring some other characteristic of the reflected probe light. For example, instead of measuring the change in intensity, it is possible to measure the change in direction, polarization or phase of the probe light. The change in phase arises from both the change in the optical constants of the sample, as already discussed, and from the displacement of the surface. To measure the phase change, various forms of interferometer have been used [10].

To illustrate some of the points just made, we show in Fig. 2 data taken for a 180 nm copper film. In this figure, the echoes marked F come about from sound pulses that are generated by the jump in stress that exists at the free surface of the film upon absorption of the pump light pulse. Because the hot electrons diffuse very rapidly, they are able to reach the back of the film before they have completely lost their energy. However, they are unable to enter the substrate.

As a result, there is a stress set up throughout the thickness of the film, and there is a discontinuity in stress at the interface between the film and the substrate. This discontinuity leads to the generation of an acoustic pulse which propagates from the back of the sample and gives rise to the series of echoes labeled B in the figure. Note that because the electrons move so fast, the pulse that starts at the back is generated at almost the same time as the pulse coming from the front.

Ultrasonic Attenuation Measurements

In picosecond ultrasonic experiments, the sound pulse that is generated usually has a complicated shape and so contains a spectrum of frequency components. To measure the ultrasonic attenuation at frequency ω , the first step is to measure the changes in reflectivity $\Delta R_1(t)$ and $\Delta R_2(t)$ due to the first and second acoustic echoes. The Fourier transforms $\Delta R_1(\omega)$ and $\Delta R_2(\omega)$ of these echoes are then calculated. The ratio $\Delta R_1(\omega)/\Delta R_2(\omega)$ can then be used to determine the attenuation at frequency ω [1]. Note that to apply this method, it is not necessary to have knowledge of the function $f(z)$ entering in Eq. 1, but one does have to know the value of the acoustic reflection coefficient at the interface between the sample film and the substrate. To measure the attenuation in a material that is transparent, a thin metallic film can be deposited on top of the sample film and used as a transducer.

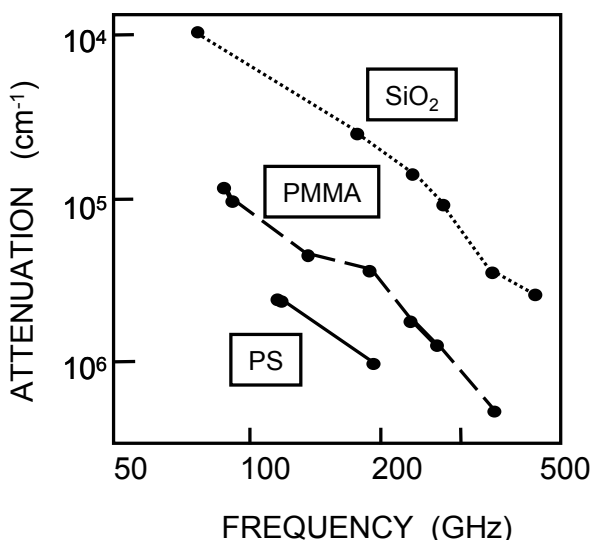


Figure 3. Attenuation of sound in SiO₂, poly-methylmethacrylate (PMMA), and polystyrene (PS) at room temperature.

Currently, only a few measurements of attenuation have been reported [1, 11, 12]. It has been found that for amorphous materials the attenuation in the frequency range 100 to 500 GHz is remarkably large. Data for polystyrene, poly-methyl-methacrylate and amorphous SiO₂ are shown in Fig. 3. At the highest frequency, the sound propagates only a few wavelengths before being attenuated.

In crystalline materials, the attenuation appears to be much smaller and is primarily due to the anharmonic interaction between the sound and the thermal phonons [13]. By lowering the temperature, the attenuation can be reduced to a small value. For example, at a temperature of 25 K or lower, it is possible to propagate sound pulses containing Fourier components as high as 200 GHz several millimeters without a large attenuation. When a short sound pulse propagates such a long distance, it is possible to observe a change in pulse shape due to the dispersion that occurs when a wave propagates through the periodic structure of the crystal lattice [14]. If the amplitude of the pulse is increased, acoustic solitons are formed [15].

Reflection of Sound at Interfaces

In ultrasonic measurements in the MHz range with piezoelectric transducers, it often happens that the transducer is not sufficiently well-bonded to the sample. This shows up as a ringing of the transducer when it is excited. This effect also occurs in picosecond ultrasonic experiments. When a thin film is deposited onto a substrate, there may be a layer of contamination at the interface. This layer is typically composed of soft organic material left on the substrate surface before the sample film is deposited. Because of the high frequencies involved in picosecond ultrasonic experiments, a contaminant layer as thin as 0.3 nm can have a significant effect on the acoustic reflection coefficient at an interface. As a consequence, picosecond ultrasonics is a powerful tool for the investigation of adhesion between films and substrates [16].

At an ideal interface the reflection of sound is determined by the acoustic impedance of the adjacent materials. High quality interfaces between semiconductors can be prepared by molecular beam epitaxy (MBE) and by using this technique, it is possible to make multilayer structures that provide a high reflection coefficient for phonons of a selected frequency. In one experiment, a reflector was made from a sequence of GaAs and AlAs layers (Fig. 4) [13]. Although the reflection coefficient at a single interface was only 0.08, the reflection coefficient of the multilayer structure at 56 GHz was 0.40. Results of an experiment using this structure are included in Fig. 4.

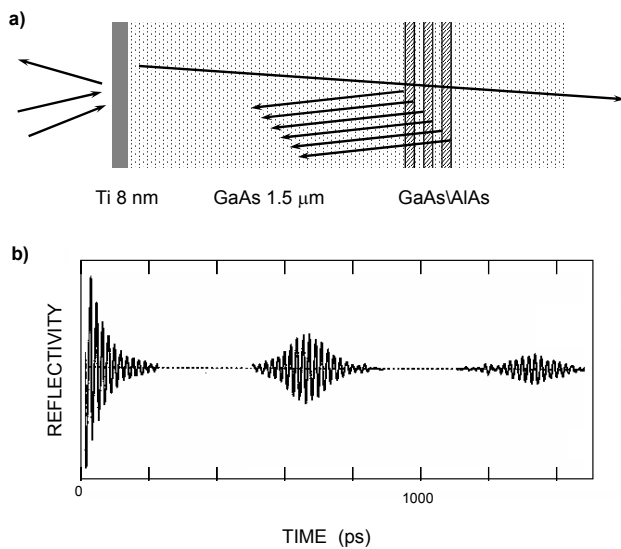


Figure 4. a) A multi-layer mirror to reflect sound of frequency 56 GHz. b) Results showing the first and second acoustic echoes from the mirror.

Vibrations of Small Structures

The discussion so far has been limited to consideration of ultrasonic experiments in thin films in which sound propagates normal to the free surface of the film and is reflected back to this surface. It is also possible to make measurements on more complicated non-planar structures [17]. Consider, for example, a structure consisting of a periodic array of metal cubes of side L on a substrate (Fig. 5), that is illuminated with a pump light pulse and then probed with a second pulse to measure $\Delta R(t)$. If L is above a certain critical value L_c that is typically around several hundred nanometers, it is useful to consider that the pump pulse leads to the generation of sound pulses at each surface of the cubes.

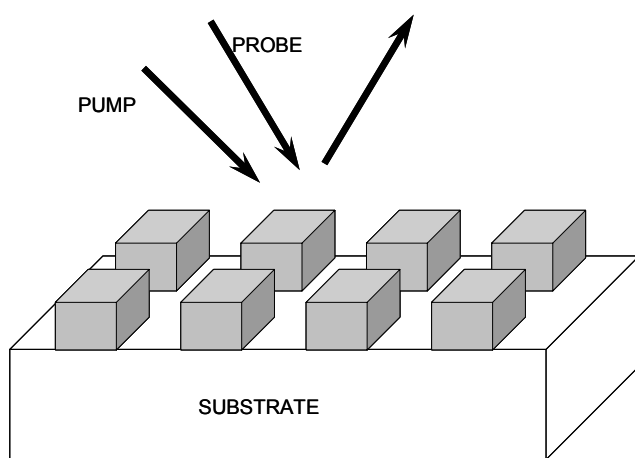


Figure 5. A sample consisting of an array of cubes on a substrate.

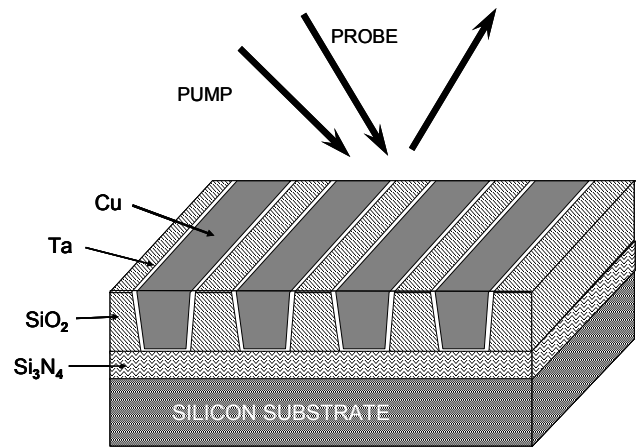


Figure 6. Sample consisting of an array of copper lines.

These pulses will reverberate within the structure, eventually damping out due to either attenuation or transmission into the substrate. Each time one of these pulses reaches a surface that can be reached by the probe light, there will be a change in the optical reflectivity $\Delta R(t)$. From the arrival time of these pulses the dimensions of the sample can be determined.

When the pump light pulse is absorbed in structures with dimensions below L_c , the excited electrons diffuse a distance that is comparable to the dimensions of the structure. As a result, the initial stress that is set up is distributed throughout most of the structure. In this situation, it is often useful to consider that the pump pulse excites different normal modes of the structure. In a first experiment of this type Lin *et al.* [18] studied an array of gold bars with dimensions 40 nm by 200 nm deposited onto a SiO₂ substrate. From the measured response $\Delta R(t)$, it was possible to determine the frequencies of two of the normal modes of the gold bars, and the vibration pattern of these two modes.

Recently, a detailed study has been made of the more complicated structure shown in Fig. 6 [19]. This consists of an array of copper wires of cross-sectional dimension 360 nm by 355 nm embedded in SiO₂. Each wire is surrounded by a "liner" of 25 nm of Ta. These dimensions of the structure were determined by electron microscopy. The goal of the experiment was to determine as many normal modes of the structure as possible and to compare the measured frequency of these modes with the results of calculations of the mode frequencies. Measurements were made on the sample in a series of experiments in which the polarization and propagation direction of the pump and probe beams were varied. It was possible to make an accurate determination of the frequency of six normal modes.

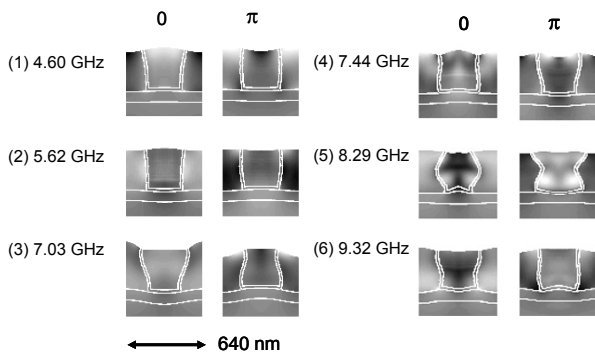


Figure 7. Displacement patterns and frequencies of the vibrational modes of the structure shown in Fig. 6.

An interesting result was that the amplitude with which a given mode appeared in the measured $\Delta R(t)$ varied by a large amount when the polarization and propagation direction were altered. Good agreement was obtained between the measured and calculated frequencies. The displacement patterns of the six modes are shown in Fig. 7.

In the two experiments just described, the pump and probe light pulses illuminate an area containing a large number of structures which, in principle, have identical geometry and elastic properties. To be able to look at individual structures, rather than an ensemble, is also of considerable interest. One can consider the following possible ways to do this:

A) The pump and probe beams illuminate a large area of the sample containing many structures, but the probe light that is reflected from just one structure is collected and sent to the detector.

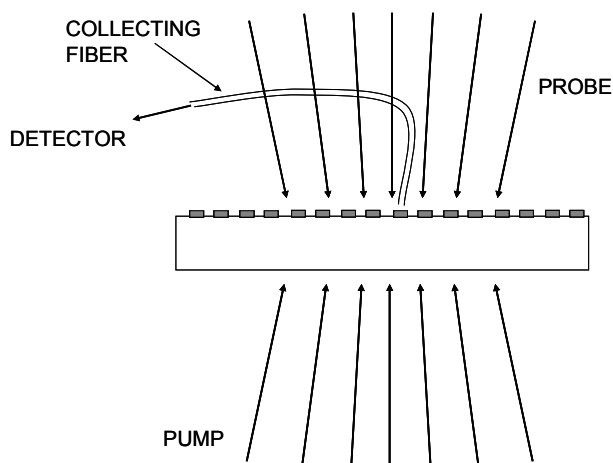


Figure 8. Schematic diagram of an experiment to study the vibrations of an individual nanostructure.

B) The pump beam illuminates a large area, but the probe light is directed so that it is incident on only a single structure.

C) The pump light pulse can be directed so that it excites only a single structure.

We have performed experiments [20] using approaches A) and B), and the set up for experiment A) is shown schematically in Fig. 8. The sample had a transparent substrate so it was possible to excite the structures with pump light passing through the substrate. For A), the probe light was focused to a spot of diameter 10-20 μm . Probe light reflected from an individual, selected, structure was collected by a fiber tip positioned close to the surface, and then brought to a Si-diode photodetector. The fiber tip was pulled down to a diameter of 100 nm and was coated with aluminum. The spatial resolution of the system was estimated to be 140 nm. For B) probe light was introduced through the fiber and directed to a single structure. To avoid damage to the tip, it was necessary to limit the power introduced into the fiber to 5 mW. The probe light reflected from the structure was collected by a lens and then directed to the photo-detector.

In the experiments on single structures, the signal-to-noise ratio is lower primarily because of the shot noise associated with the smaller photon flux on the detector. As a result, it is necessary to perform more signal averaging and the time needed to make a measurement is longer.

Practical Applications

In the computer chip industry, it is of great importance to be able to measure and control the thickness of thin films. Transparent films can be measured using ellipsometry, but this technique cannot be applied to opaque films. Before the development of the picosecond ultrasonics technique, it was not possible to make rapid and non-destructive measurements of the thickness of metal films. Rudolph Technologies [21] has developed a sophisticated instrument that uses picosecond ultrasonics to make measurements of the thickness of films in a computer chip and other microelectronic devices. For operation in an industrial environment, the instrument has to be completely automated, i.e., the pump and probe beams have to be directed to selected locations on the surface of the wafer, correct focusing has to be maintained, etc. From a measurement of the change in optical reflectivity with time, the instrument is able to determine the thickness of each layer in a stack of several thin films of different materials and thickness. A structure of this type is shown in Fig. 9 along with the data that is obtained [22]. The features in $\Delta R(t)$ observed in the

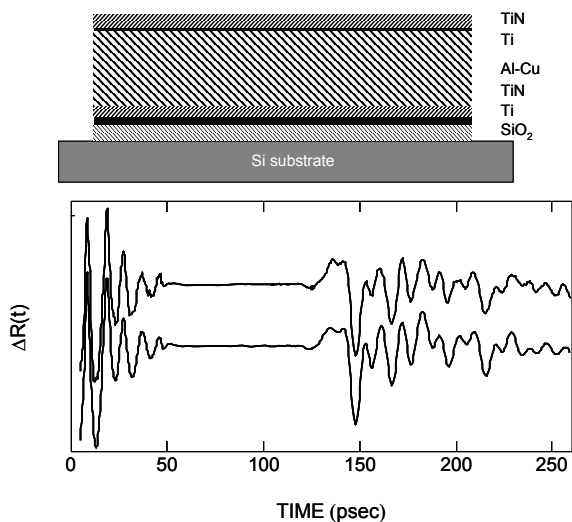


Figure 9. Multilayer structure together with experimental data (upper curve) and fit from computer simulation (lower curve). Results taken from ref. 22 .

first 50 ps arise from the sound bouncing back and forth in the top two layers of the structure (Ti and TiN). The features after 130 ps come from sound that has made one round trip across the thick Al-Cu layer in the middle of the structure.

To determine the thickness of all of the films from the measured data $\Delta R_{\text{exp}}(t)$, a trial guess at the thickness is made and then a computer simulation is performed to determine the expected $\Delta R_{\text{sim}}(t)$. The thickness of each film is then adjusted to give the best agreement between $\Delta R_{\text{exp}}(t)$ and $\Delta R_{\text{sim}}(t)$. It can be seen from Fig. 9 that a very detailed fit to $\Delta R_{\text{exp}}(t)$ can be obtained, and this make it possible to determine an accurate value for the thickness of each of the six films in the film stack.

Summary

The advent of pulsed lasers that can produce pulses of duration less than 1 ps has made ultrasonic measurements possible on structures in which the acoustic path is 10 nm or even less, and has greatly extended the maximum frequency at which ultrasonic measurements can be made.

Acknowledgments

We acknowledge contributions to this research by many students in our laboratory, particularly G.A. Antonelli, H.-Y. Hao, C.J. Morath, R.J. Stoner and G. Tas. This work was supported in part by the US Air Force Office of Scientific Research MURI grant entitled "Phonon Enhancement of Electronic and Optoelectronic Devices," (Grant No. F4962-00-1-

0331), through a partnership award from IBM, and by the Intel Research Council.

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