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Ultrafast acoustics for imaging at the nanoscale

B C Daly[1] **and T B Norris**[2]

[1] Physics and Astronomy Department, Vassar College, Poughkeepsie, NY 12604
[2] Center for Ultrafast Optical Science and EECS Department, University of Michigan, Ann Arbor, MI, 48109

E-mail: brdaly@vassar.edu

Abstract. In this paper we present a series of experiments which show that 2-D and possibly 3-D imaging with sub-micron resolution is possible by means of ultrafast acoustic techniques. Optical pulses from a Ti:sapphire laser are used to generate picosecond acoustic pulses on one side of a ~1 mm thick Si wafer. The 1 mm distance is sufficient for the acoustic waves to diffract to the far field before they are detected by time-delayed probe pulses from the Ti:sapphire laser. The acoustic waves are either generated by a surface nanostructure or scattered from a buried nanostructure, and an image of that nanostructure is reconstructed through an analysis of the detected acoustic waves.

1. Introduction

Picosecond ultrasonics was first studied roughly twenty years ago by Maris' group at Brown University [1] and by Eesley's group at Ford Motor Company. [2] The well-known principle behind this method is that sub-picosecond optical pulses can generate and detect longitudinal acoustic pulses with frequencies of a few 100 GHz in thin metal films. This coherent phonon generation and detection method has proven to be extremely useful for a wide range of investigations into the physics of thin films, as well as for 1-D thin film thickness metrology. More recently, we have shown that 2-D picosecond ultrasonic imaging with sub-micron resolution is possible. [3] This was achieved by modifying the traditional picosecond ultrasonics arrangement so that the acoustic phonons are generated on one side of a 1 mm thick Si wafer but detected on the far side of that wafer. The resulting measurement allows for detection of the acoustic phonons in the far field of the source. In this paper we describe two measurements. In the first measurement, patterned Al transducers were used to generate the acoustic waves, and in the second measurement, a buried nanoscale photoreseist pattern was used to scatter the acoustic waves. Images of those structures with sub-micron resolution were obtained via a reconstruction algorithm that accounts for the elastic anisotropy of the Si wafer.

2. Imaging of surface nanostructures

We used ultrafast optical generation and detection of single-cycle, 100 GHz bandwidth coherent acoustic phonon pulses for nanometer scale imaging. The concept underlying this imaging method has previously been demonstrated for both 2-D [4] and 3-D [5] imaging with single-cycle terahertz electromagnetic pulses. Far field time-domain measurements of waves scattered from an object can be numerically back-propagated to produce a reconstruction of the original wave field at the object plane, and therefore of the object itself.

As a first experiment we attempted to make acoustic images of an Al pattern grown on a Si wafer. The counter-propagating optical pump-probe setup is shown in figure 1 and a close-up of the sample is shown in figure 2. The sample was a double-side-polished, high-purity, [111] Si wafer (thickness = 0.5 mm) that was coated on both sides with thin (15 nm) films of Al. 1 μm width lines were etched into the Al film on one side using e-beam lithography techniques. The experiment is arranged so that the pump pulses were incident on the patterned transducer and the probe pulses are incident on the non-patterned transducer, directly opposite the pump spot. The ultrafast optical source was a Ti:sapphire laser with a repetition rate of 76 MHz. A rapid delay stage was included in the pump arm of the experiment to allow for signal averaging over a time delay window of ~150 ps. A second stage with a length equal to that of the laser cavity was included so that arbitrary delays (longer than the repetition time $\tau = 13.2$ ns) could be sampled. This was necessary due to the fact that the acoustic pulses arrived at the probe transducer at a delay time greater than τ. The pump beam was acousto-optically modulated at a frequency of ~ 1 MHz to enable lock-in detection of the probe reflectivity. The sample was held at a temperature below 20 K in a He flow cryostat in order to suppress phonon-phonon scattering and thereby allow phonon propagation over distances of a few mm. A nearly diffraction-limited optical probe spot was maintained during the measurements by using a microscope objective that was external to the cryostat. We measured the scattered acoustic field by scanning the probe microscope objective over an array of off-axis detection positions separated by 1 μm.

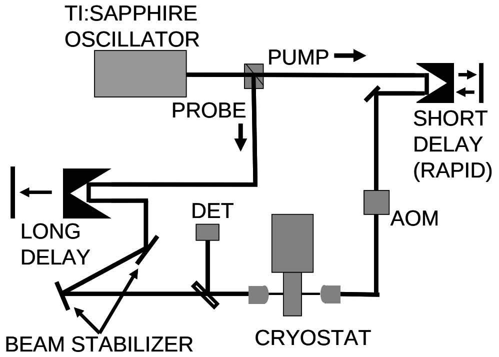

Figure 1. Optical pump-and-probe setup

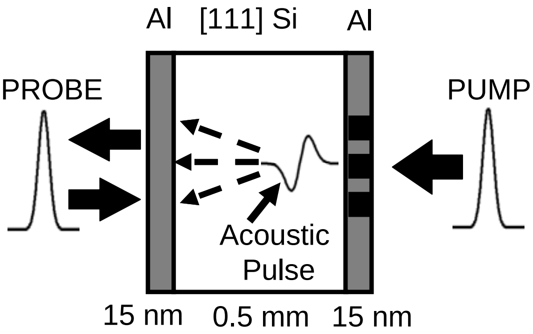

Figure 2. Schematic of the sample in a surface acoustic imaging experiment

For this experiment, the energy of the pump pulses was typically 0.1 nJ, and this energy was distributed over a spot that had a Gaussian profile with a radius of about 3 μm. The resulting thermal expansion of the Al produced roughly single-cycle longitudinal acoustic phonon pulses that propagated into the film and substrate. Since the heated Al film was patterned, the acoustic field that was launched into the substrate had the same shape as the pattern itself. It is important to note that in this experiment the object to be imaged was actually the source of the detected waves as opposed to a scatterer of the detected waves. For the geometry shown in figure 2 the acoustic phonons had a peak frequency and bandwidth of ~100 GHz, and the acoustic strain amplitude was of the order of 1×10^{-5}. As the acoustic phonons propagated to the far side of the Si wafer, they diffracted sufficiently so that by the time they reached the far side of the wafer, spatial variations in the acoustic field were large enough to be probed by a 1 μm diameter optical probe beam.

At each detection point, the slight change in reflectivity induced by the time-varying acoustic field was used to determine the acoustic field amplitude. When the probe beam was located on the propagation axis, the detected change in reflectivity, ΔR was that of the usual single cycle acoustic pulse. For off-axis positions however, the data appeared as in figure 3, which shows ΔR versus time delay for a particular probe spot located 20 microns off axis in a direction perpendicular to the lines. It is important to note that the delay time shown has been shifted by the ~52 ns that it takes for an individual acoustic pulse to traverse the 0.5 mm Si wafer. The data show a series of peaks that are separated by a few ps. These peaks are indicative of the acoustic waves arriving from the different lines illuminated by the pump spot. It is our ability to temporally resolve these peaks that allows us to numerically reconstruct the object that launched the waves.

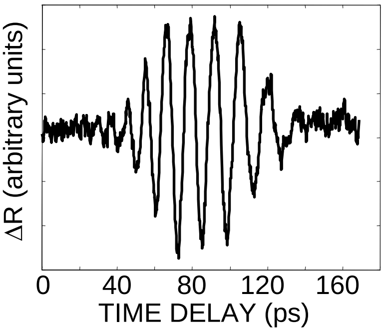

Figure 3. Reflectivity change versus time delay for a detection point 20 μm off axis on the far side of the Si wafer from the pattern. The time delay has been shifted by 52 ns to account for the time it takes for an acoustic pulse to traverse 0.5 mm of Si.

In developing the reconstruction algorithm, we began with a time-reversed diffraction integral

$$u(P_1,t) = -\frac{1}{4\pi c}\iint\left(\frac{1+\cos(\vec{n}_0,\vec{r}_{01})}{r_{01}}\right)\frac{\partial}{\partial t}u\left(P_0, t+\frac{r_{01}}{c}\right)d\sigma', \tag{1}$$

which holds for waves in an isotropic medium. Here, the double integral over $d\sigma'$ represents a summation over a surface in the far field of the wave source. $u(P_1,t)$ represents the reconstructed wave field at a point P_1 on the object plane, c is the wave speed, $\vec{n}_0$ is the normal to the far-field surface, $\vec{r}_{01}$ is the vector from point P_1 in the object plane to a detection point P_0 on the far field surface. Of course, unlike electromagnetic waves propagating in free space, acoustic waves propagating in an anisotropic crystal have a group velocity that depends on the propagation direction. The group velocity vector in a given direction is the gradient of a surface of constant frequency in wave vector space. [6,7] In practice, we included this 'phonon focusing' effect by first using the elastic constants of Si to obtain a map of the group velocities as a function of different crystallographic directions. We then compared those directions to the specific directions probed in a particular set of data, and assigned the correct group velocities for every detection point-object point pair. Figure 4 shows an acoustic image of a surface nanostructure along with corresponding SEM image.

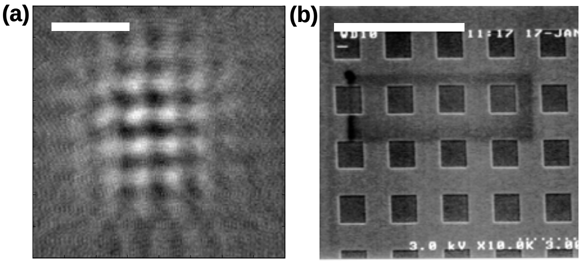

Figure 4. a) SEM image of Al grid. b) Acoustic phonon image of Al grid. The white scale bar on both plots represents a length of 5 μm (Note: the scale of a) and b) are not equal).

3. Imaging of sub-surface nanostructures

We have performed an additional experiment in order to demonstrate sub-surface acoustic imaging (Figure 5a). A 200 nm layer of UV sensitive photoresist (PR) was deposited on a 1 mm thick silicon substrate. The photoresist was exposed with 1 micron line and grid patterns identical to the patterns of the previous experiment, but the resist was left undeveloped. The result of this is that a pattern of cross-linked and uncross-linked polymer is left embedded in the PR film. We then coated both the PR and bare sides of the Si wafer with 15 nm Al film transducers. Acoustic phonon pulses were launched through the latent PR pattern. As in the previous arrangement, the acoustic field was measured at a series of probe positions at the Al film on the far side of the wafer. As a result of the roughly 10% increase in the sound velocity in the UV-exposed PR, the acoustic field that is measured is that of two identical, but laterally shifted patterns that arrive roughly 8 ps apart. The reconstruction algorithm is designed to produce a 2-D image of the scattering object by reproducing the spatial profile of the field at one particular time. In figure 5b we use a different type of reconstructed image for the case of a 1 μm line pattern in the photoresist. The x-axis shows position in a direction perpendicular to the lines, but the y-axis now represents different backpropagation times. Thus, figure 5b is a stack of 1-D lineouts of the reconstructed image for different backpropagation times. The image shows two sets of 1μm lines that are shifted by 8 ps. Figures 5c and 5d are the 2-D images at the times indicated by the dotted black lines shown on figure 5b.

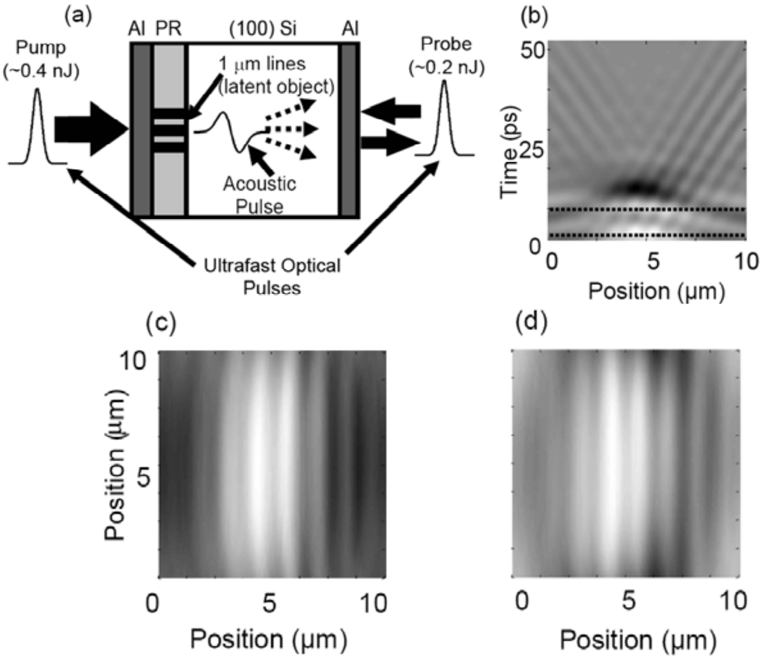

Figure 5. Latent photoresist pattern experiment a) Setup. b) Stack of 1-D reconstructions as a function of time. (c) 2-D image at 0 ps (d) 2-D image at 8 ps (delay times shifted by ~ 118 ns)

In summary, we have described two experiments that indicate that surface and sub-surface imaging of nanostructures is possible using a modification of the traditional picosecond ultrasonics experiment. Our next step will be to begin work on improving the resolution towards the phonon wavelength-limit which is expected to be better than 50 nm.

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