

1 **Picosecond ultrasonic study of surface acoustic waves on periodically
2 patterned layered nanostructures**

3 Michael Colletta,^a Wanjiru Gachuhi,^a Samuel A. Gartenstein,^a Molly M. James,^a Erik A. Szwed,^a
4 Brian C. Daly,^{a†} Weili Cui,^b George A. Antonelli,^c

5 a: Vassar College, 124 Raymond Ave, Poughkeepsie, NY, USA 12604

6 b: SUNY Maritime College, Throggs Neck, NY, USA 10465

7 c: Antonelli Research and Technology, 3227 SE Lambert St, Portland, OR, USA 97202

8 †: corresponding author's email: brdaly@vassar.edu

9 **Abstract:**

10 We have used the ultrafast pump-probe technique known as picosecond ultrasonics to generate
11 and detect surface acoustic waves on a structure consisting of nanoscale Al lines on SiO₂ on Si.
12 We report results from ten samples with varying pitch (1000 nm to 140 nm) and SiO₂ film
13 thickness (112 nm or 60 nm), and compare our results to an isotropic elastic calculation and a
14 coarse-grained molecular dynamics simulation. In all cases we are able to detect and identify a
15 Rayleigh-like surface acoustic wave with wavelength equal to the pitch of the lines and
16 frequency in the range of 5 GHz – 24 GHz. In some samples, we are able to detect additional,
17 higher frequency surface acoustic waves or independent modes of the Al lines with frequencies
18 close to 50 GHz. We also describe the effects of probe beam polarization on the measurement's
19 sensitivity to the different surface modes.

20 **Keywords:** ultrafast optics; picosecond ultrasonics; laser ultrasonics; surface acoustic waves

21 **Introduction**

22 The highest frequency surface acoustic waves (SAWs) have to date been generated and detected
23 by ultrafast optical pump-probe techniques.^{1,2,3,4} The most common method is to use lithography
24 to fabricate a nanoscale pattern of metallic lines on a substrate and excite those lines by means of
25 an ultrafast laser pulse. The rapid thermal expansion of the lines launches a SAW (often called a
26 pseudo-SAW or leaky SAW since the presence of the pattern on the surface causes radiation of
27 acoustic energy into the bulk) with wavelength equal to the pitch of the lines that propagates
28 perpendicular to the lines. SAWs up to 50 GHz have been generated and detected in this
29 manner.⁵ In recent work conducted by two authors of the present article we measured SAWs or
30 related surface vibrations in a complicated structure consisting of TiN wires of nanometer scale
31 cross-section grown on a multilayered stack of porous and non-porous oxides on an Si wafer.⁶
32 These unique samples yielded pitch-dependent frequencies that in some cases compared
33 favorably with Rayleigh-like or Sezawa-like surface waves,^{7,8} but in other cases corresponded to

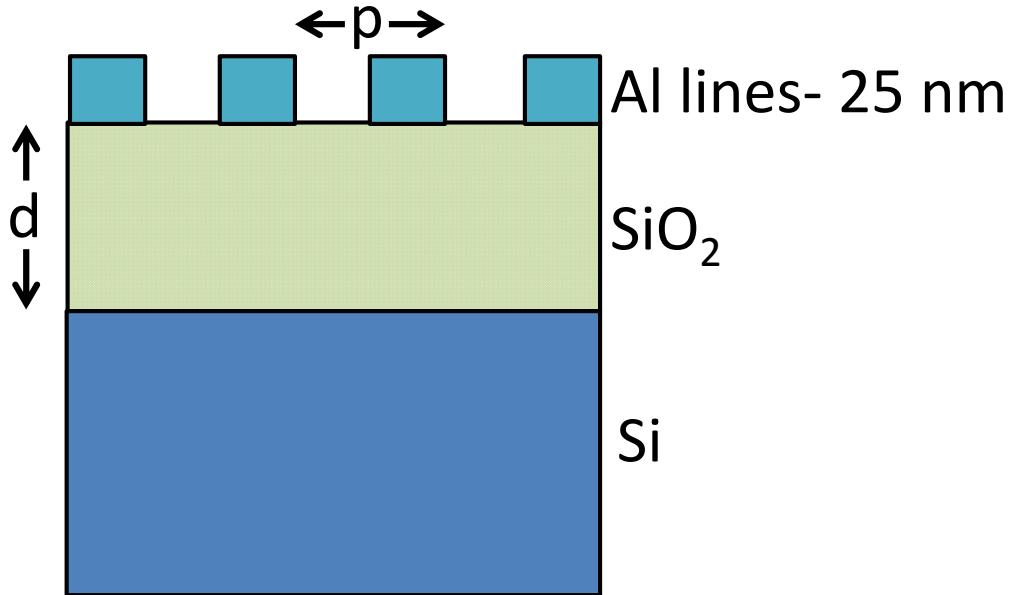
1 modes that radiated significant acoustic energy into the substrate. In this work we have chosen
2 to study a simpler set of samples consisting of Al lines on thin SiO_2 on Si and we detect multiple
3 SAWs which can be identified by comparison with coarse-grained molecular dynamics
4 simulations. The number of modes detected was found to depend on the pitch of the patterned
5 Al as well as on the wavelength and polarization of the probe light. We detected Rayleigh-like
6 SAWs and Sezawa-like SAWs with wavelength equal to the pitch of the Al lines as well as
7 SAWs with wavelength equal to one-half or one-third of the pitch.

8 Experiment

9 The ultrafast optical pump-probe experiment known as picosecond laser ultrasonics (PLU) has
10 been described extensively in the literature.^{9,10} We performed this experiment with a Ti:Sapphire
11 oscillator operating at a 76 MHz repetition rate with pump wavelength of 800 nm and probe
12 wavelengths of 800 nm or 400 nm. The 400 nm probe experiments were achieved by second
13 harmonic generation in a BiBO crystal. The 10 patterned samples that we studied are illustrated
14 schematically in Fig. 1. The samples were fabricated at the Cornell NanoScale Facility by the
15 following process: thermal oxidation to produce the amorphous SiO_2 layer of thickness $d = 60$ or
16 112 nm, thermal evaporation of 25 nm of Al, and E-beam lithography and dry etching to create
17 the nanometer scale Al pattern. The lines were etched perpendicular to the [110] direction in the
18 Si substrate, they varied in pitch p ranging from 140 nm up to 1000 nm, and they were all etched
19 near 50% (ranging from 40-60%) duty cycle. The patterned samples were placed into the optical
20 setup, where pump and probe beams were both focused onto the same 20 μm diameter spot, so
21 that anywhere from 20 to 140 periods of the pattern were strongly illuminated. For the data
22 reported here, the pump and probe beams were held normally incident. Measurements were also
23 made with 45 degree incident probe light but only minor differences from the normal case were
24 found. The ultrafast pump pulses were absorbed by the Al lines, and the resulting rapid thermal
25 expansion launched ultrasonic waves both downward into the SiO_2 film and Si substrate, and
26 laterally as SAWs in the direction perpendicular to the line pattern. The various ultrasonic waves
27 can be detected by the time-delayed probe pulses due to transient changes in the reflectivity ΔR
28 that they cause. The sources of these transient changes include the dependence of the optical
29 constants of the Al on strain as well as the changes in reflectivity of the optical grating produced
30 by the nanostructure as it responds to the acoustic oscillations. In this work, we focus on the
31 signals caused by laterally propagating ultrasound and not the signals caused by acoustic waves
32 traveling normal to the sample surface.

33 Fig. 2a shows an example of the ΔR signal with 800 nm probe as a function of probe delay time
34 for two of the samples ($p/d = 400$ nm/112 nm and 200 nm/112 nm). The exponentially decaying
35 thermal background and the initial jump at $t = 0$ have been subtracted off so that the dominant
36 oscillations are easier to observe. Fig. 2b shows the Fourier transform for these two data sets.
37 For both samples, the data are dominated by oscillations at two frequencies. As is expected, the

1 **Figure 1**



2

3

4 Figure 1: Schematic diagram of the samples. Film thickness d was either 60 nm or 112 nm as
5 measured by picosecond ultrasonics. Al lines pitch p varied from 1000 nm down to 140 nm.
6 The duty cycle was close to 50% (+/- 10%) in all cases as measured by SEM imaging.

7

8

9

10

11

12

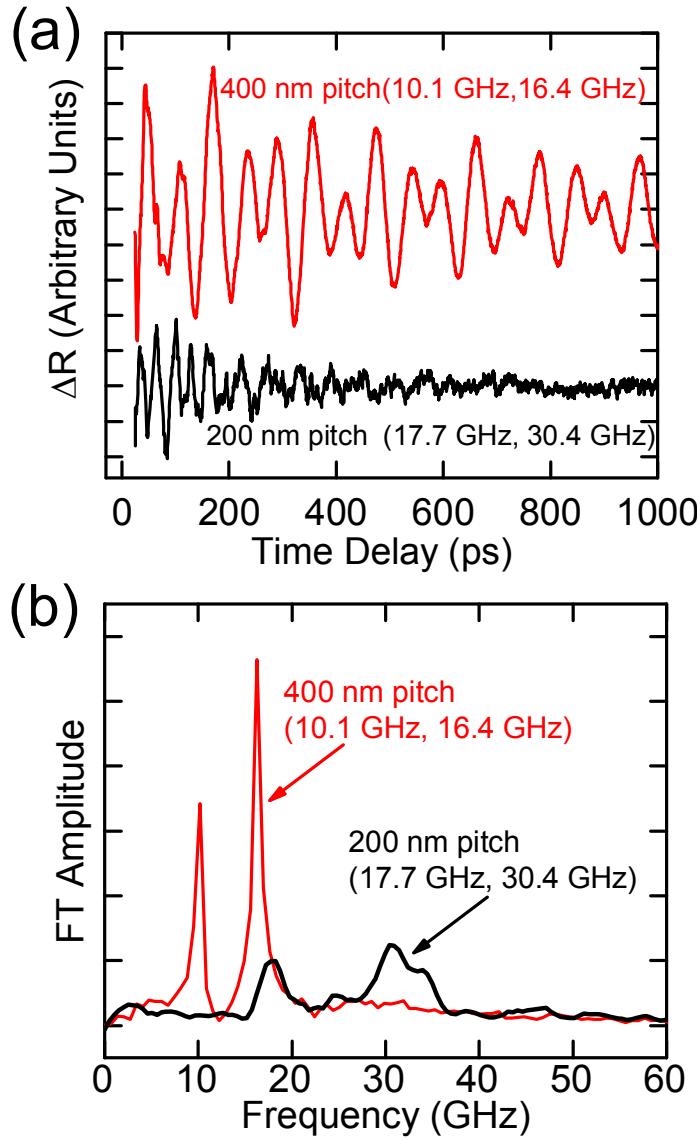
13

14

15

16

1 **Figure 2**



2

3 Figure 2: (a) ΔR for two samples with $d = 112$ nm and $p = 400$ nm and 200 nm. (b) Fourier
4 transform amplitude of the signals in (a).

5

6

7

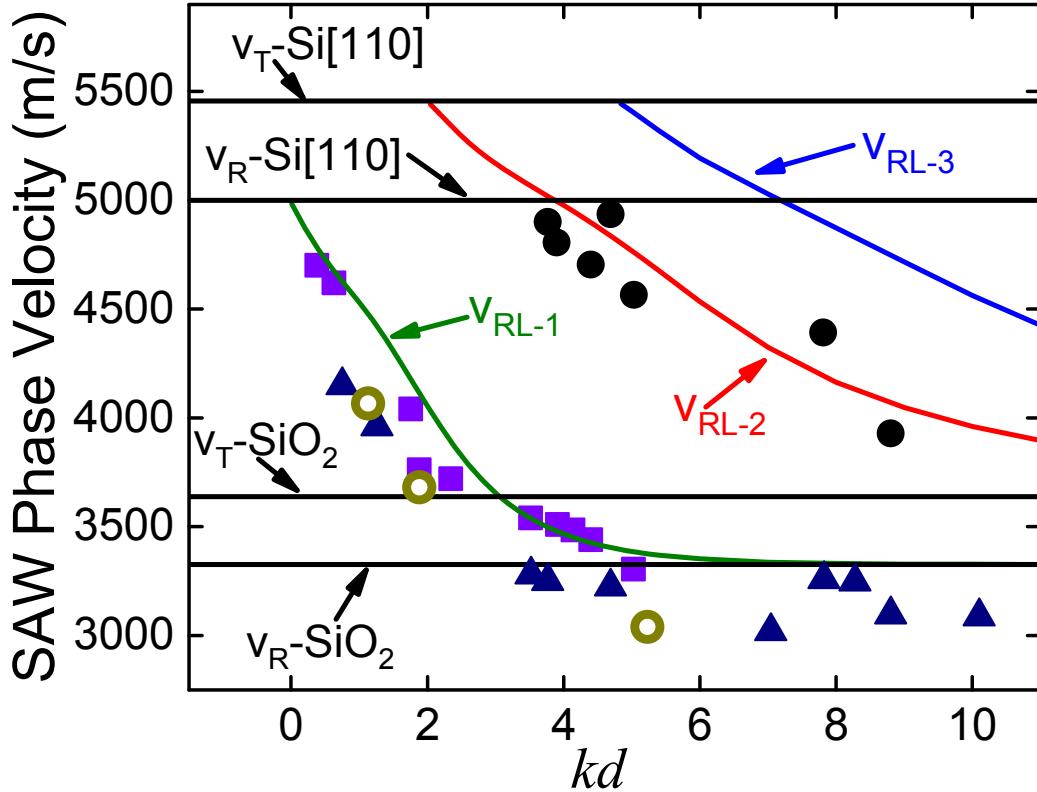
1 smaller pitch sample produces higher frequency oscillations. The frequencies of the observed
2 oscillations are strongly dependent on the pitch of the samples, and so it is evident that they must
3 represent SAWs, independent bar modes, or a wave propagating very near to the surface of the
4 sample. Other data sets show as few as one or as many as seven detected surface modes, and in
5 the following sections we identify the nature of many of these modes. It is also evident from Fig.
6 2a that the excitations are shorter lived in the 200 nm sample as compared with the 400 nm
7 sample. The presence of a periodic structure on a surface adds stress which in turn causes
8 radiation of surface acoustic wave energy into the bulk - a process that was first described by
9 Brekovskikh.¹¹ This is the dominant damping mechanism for SAWs on periodically patterned
10 surfaces in the GHz range, and the damping depends on the fourth power of the SAW
11 frequency.^{4,12}

12 **Lowest order Rayleigh-like modes**

13 The lowest frequencies measured for all ten samples compare favorably with the expected
14 frequency for a Rayleigh-like wave in Si in the [110] direction that is loaded by a thin oxide
15 layer. Table I lists the frequencies of the lowest frequency oscillations detected for the 10
16 samples. In the only case where we measured two films with the same pitch but differing oxide
17 thickness ($p/d = 200\text{nm}/60\text{nm}$ and $200\text{nm}/112\text{nm}$) we clearly see that the effect of the thicker
18 oxide is to lower the frequency of the detected wave. This is expected for this situation where
19 the SiO_2 film loads the Si substrate due to its lower density and sound velocity. Fig. 3 contains a
20 plot of the phase velocities for the 10 measured Rayleigh-like SAWs alongside an analytical
21 calculation of the phase velocities for an isotropic model of an SiO_2 loaded Si substrate.⁸ The x-
22 axis of the graph is the dimensionless quantity kd where k is the wavenumber of the SAW. Note
23 that the calculation of the first Rayleigh-like mode has the Rayleigh velocity of Si for very small
24 kd , but has the Rayleigh velocity of SiO_2 for very large kd . The data are in good agreement with
25 the predicted velocities (deviating by between 0.7% and 8%) which is reasonable given that the
26 experimental uncertainty in the measurement of the frequencies is 5%. The plot also shows that
27 for a case such as this one, in which the thin film loads, rather than stiffens, the surface of the
28 substrate, additional modes (labeled RL-2 and RL-3 in Fig. 3) appear as kd is increased. The
29 RL-2 mode is also called a Sezawa wave.

30 To further confirm the nature of the SAWs that we detected, we have performed 2D coarse-
31 grained molecular dynamics (CGMD) simulations on cross-sectional models of the
32 nanostructures. The simulations essentially collect multiple atoms together to form larger super-
33 atoms in order to solve the elastic motion of the cross section of the structures in question. The
34 simulation, which has been described in detail previously,^{6,13} models the materials as isotropic
35 elastic solids with given density and transverse and longitudinal sound velocities. We simulate a
36 2D cross-section of one period of the structure, and implement periodic boundary conditions in
37 the lateral directions. We also place an absorbing boundary condition at the bottom of the
38 structure so that any simulated modes traveling into the substrate will not be reflected back to the

1 **Figure 3**



2
 3 Figure 3: Phase velocity v of the lowest frequency mode detected on the ten samples (■) as
 4 determined by the formula $v = f\lambda$ where λ = pitch p . The calculated phase velocity for the first
 5 three Rayleigh-like surface wave modes for isotropic SiO₂ on Si are plotted with the green, red,
 6 and blue solid curves respectively. The (▲) and (○) symbols represent the measured frequencies
 7 that correspond to the RL-1 modes for the $\lambda = p/2$ and $p/3$ cases, respectively. The RL-2
 8 (Sezawa) modes identified in the experiment and reported in Table II are plotted as well (●).
 9 The transverse velocity and Rayleigh wave velocity for Si [110] and SiO₂ are represented by the
 10 solid black lines.

11
 12
 13
 14

1 **Table I.**

Sample (p/d in nm)	Freq. Meas. at 800 nm (GHz)	CGMD simulation (GHz)	IEC Calculation (no Al bars) (GHz)
1000/60	4.7	4.8	4.8
600/60	7.7	7.7	7.8
400/112	10.1	10.2	10.4
300/112	12.4	12.6	13.0
200/112	17.7	17.7	17.8
200/60	18.8	19.9	20.6
180/112	19.5	19.1	19.4
170/112	20.5	19.9	20.3
160/112	21.5	21.1	21.5
140/112	23.6	23.4	24.1

2

3 Table I: Column two contains the lowest measured frequency for the 10 samples in the study
 4 using the 800 nm probe configuration. Column three contains the CGMD simulated frequency
 5 of the RL-1 mode with $\lambda = p$ from column one. Column four contains the frequency of the RL-1
 6 mode with $\lambda = p$ calculated using the methods of Ref. 8 for an isotropic elastic continuum (IEC)
 7 of SiO_2 on Si (with no Al bars).

8

9

10

11

12

13

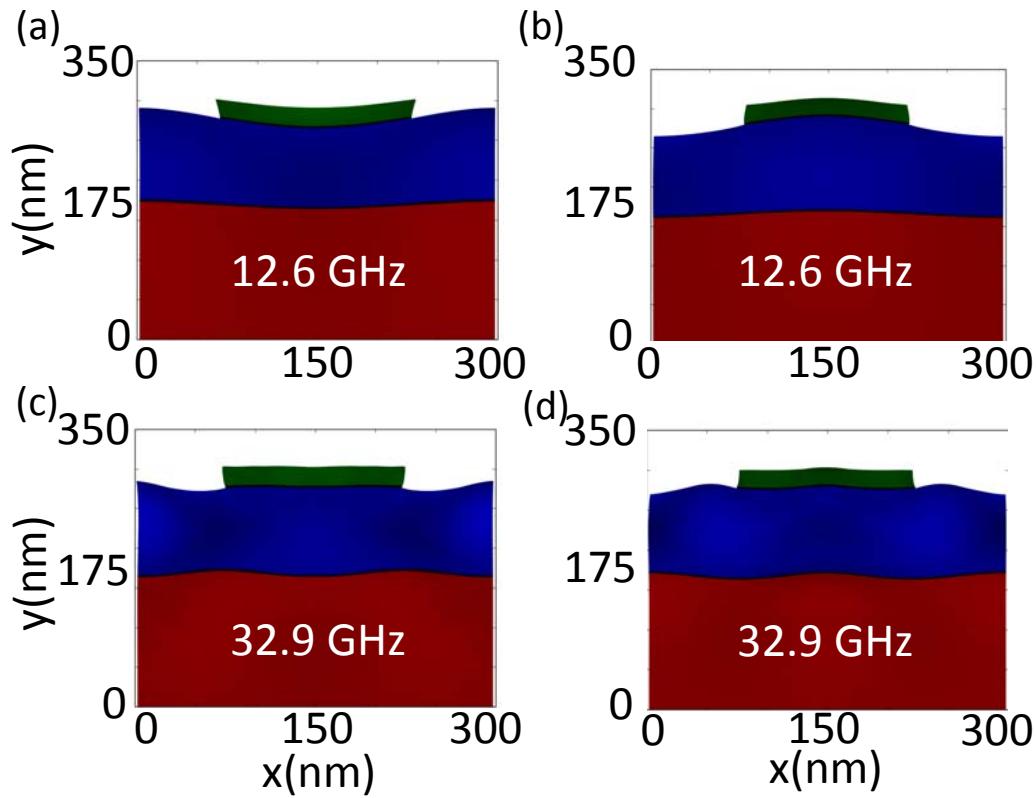
1 surface. The simulation has no other source of damping, so unlike in the experiment, any pure
2 surface mode that is excited will remain excited indefinitely. To simulate the pump-probe
3 experiment, we heat the Al bars instantaneously in order to excite the possible modes of the
4 structure. We then plot the Fourier transform of the displacement versus time over a range of a
5 few thousand ps at various locations in the model. Once we have determined what the dominant
6 frequencies are, we run the simulation a second time, this time driving the model at a particular
7 frequency so that a visualization of each mode can be created. The simulations allow us to
8 observe the perturbative effects of the presence of the 25 nm thick Al lines on the modes
9 available to the structure. The frequencies of the lowest order Rayleigh-like mode provided by
10 the simulation are given alongside the measured frequencies in column three of Table I and show
11 a slightly improved agreement (maximum deviation of 5%) with the data as compared to the
12 analytical calculation (column four). More importantly, the simulation allows us to visualize the
13 modes to ensure that the motion is consistent with a Rayleigh-like SAW. Figures 4a and 4b are
14 two frames of an RL-1 wave simulation for the $p/d = 300\text{nm}/112\text{nm}$ sample driven at 12.6 GHz.
15 The characteristic form of the RL-1 wave is that the displacement of the top and bottom of the
16 SiO_2 layer are in phase with one another (the film is flexing). We have successfully modeled
17 (with frequency matching to within 10% in all cases, better than 5% in most cases) nearly every
18 one of the vibrational modes detected in the pump-probe experiment, including many of the
19 higher frequency modes described in the following section. The simulations were executed with
20 a precision of ± 0.2 GHz.

21 **Higher frequencies**

22 The pump-probe experiments have also shown a number of higher frequencies above the 1st
23 order Rayleigh-like mode. This is in contrast to several similar experiments found in the
24 literature, including our work of Ref. 6, but it is expected given the higher order modes
25 illustrated by the solid curves of Fig. 3. In a 2009 paper, Maznev and Every calculated the
26 Brillouin zone-folded SAW band structure for a sample whose structure is similar to the ones
27 measured here. Their work showed that the presence of a periodic array of lines atop a film
28 coated substrate produces hybridization of the first Rayleigh band with the Sezawa band, and
29 that the expected frequencies are shifted from the case of a uniform film due to avoided crossings
30 of these bands.¹⁴ We also find a dependence on the wavelength of the probe light and the
31 polarization of the probe light relative to the orientation of the lines.

32 Table II gives all of the frequencies measured on the 10 samples at the two probe wavelengths
33 and for probe beam polarizations perpendicular (S-polarized) and parallel (P-polarized) to the
34 orientation of the Al lines. The pump excitation of the nanostructure can excite SAWs with
35 wavelength $\lambda = p, p/2, p/3 \dots$. If the periodicity of the structure possessed a perfect 50% duty
36 cycle we would not generate the even modes in this series, but we do observe the 2nd mode in
37 every sample, even though the duty cycles as measured by SEM deviated from 50% by 10% or
38 less. The measured frequencies that we associate with the $\lambda = p/2$ and $p/3$ RL-1 modes are

1 **Figure 4**



2

3 Figure 4: Coarse-grained molecular dynamics visualizations of two vibrational modes for the
4 $p/d = 300\text{nm}/112\text{nm}$ sample. Displacements are greatly exaggerated so that the character of the
5 mode can be observed. (a) and (b) are two frames of the visualization for a 12.6 GHz RL-1 mode
6 with $\lambda = 300$ nm that is detected in the experiment at 12.3-12.4 GHz. (c) and (d) are two frames
7 of the visualization for a 32.9 GHz RL-2 mode with $\lambda = 150$ nm that is detected in the
8 experiment at 32.9 GHz.

9

10

11

12

13

14

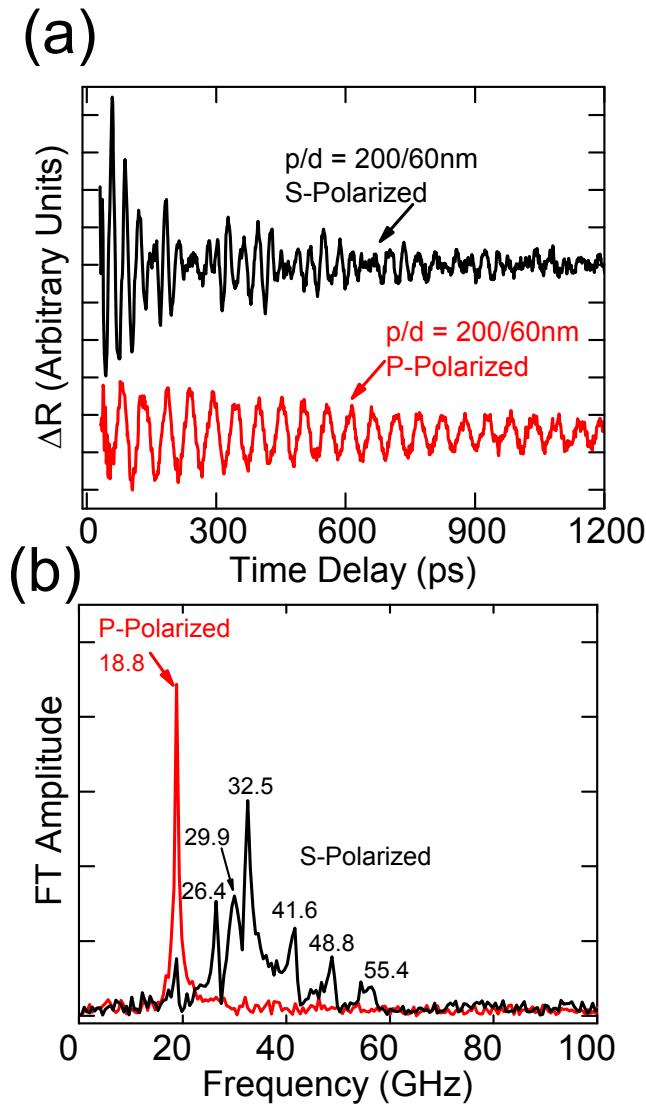
15

1 plotted alongside the lowest frequency modes in Fig. 3. Within experimental uncertainty,
2 several of these modes have a measurably lower frequency and phase velocity than is predicted
3 by the isotropic elastic continuum calculation - as much as 15% lower. Frequency shifts in the
4 Rayleigh-like wave frequency away from the pure Rayleigh wave frequency are well known for
5 periodic arrays of lines,^{4,5,15} and the magnitude of the effect can be calculated by perturbation
6 theory and is proportional to the ratio of the line thickness to the pitch, which is $25\text{nm}/p$ in our
7 case. Since our data do not clearly demonstrate the pitch dependence of this effect, nor
8 demonstrate the effect at all for the lowest frequency RL-1 modes, this remains for us a topic of a
9 future study.

10 In addition to RL-1 modes, Table II also includes the RL-2 modes (also plotted in Fig. 3) and a
11 few individual bar modes which is indicative that some of our data sets are very rich in
12 frequencies. Fig. 5 shows the reflectivity data for 400 nm probe on the $p/d = 200\text{ nm}/60\text{ nm}$
13 sample and its Fourier transform. When the probe light is P-polarized, only a single frequency is
14 readily visible in the data, but when the probe light is S-polarized, seven frequencies are easily
15 distinguished. The 18.8 GHz peak (which is relatively weak for S-polarized probe) is readily
16 identifiable as the lowest frequency RL-1 wave, similar in shape to Figs. 4a and 4b. The
17 strongest peak at 32.5 GHz differs only slightly from a small peak in the simulation at 35.3 GHz
18 which corresponds to the $\lambda = p/2 = 100\text{ nm}$ RL-1 mode. The 29.9 and 48.8 GHz peaks compare
19 well with a pair of modes from the simulation at 30.7 and 51.2 GHz which are RL-2 (Sezawa-
20 like) modes with $\lambda = p = 200\text{ nm}$ and $\lambda = p/2 = 100\text{ nm}$ respectively. The RL-2 modes have a
21 characteristic breathing motion of the film like the simulation plotted in Figs. 4c and 4d. The
22 phase velocity of the first of these two modes is higher even than the shear velocity in Si, and is
23 predicted by the simulations to be a very short-lived mode. The 26.4 and 55.4 GHz modes are
24 comparable to two modes from the simulation (27.8 and 55.4 GHz) that we have identified as
25 individual modes of the Al lines themselves as opposed to a collective motion of the whole
26 structure. Previous studies of this sort have also measured independent bar modes alongside
27 SAWs,^{1,2} and we discuss these modes later in this section. The remaining mode identified in Fig.
28 5 at 41.8 GHz (marked U in Table II for ‘unidentified’) is weakly predicted in the CGMD
29 simulations at 42.6 GHz and is possibly caused by the longitudinal head wave propagating near
30 the surface in the Si. A signal due to just such a wave was detected in the experiment of Miao et.
31 al. in which a transparent mask was brought very near to a substrate and an ultrafast pump-probe
32 scheme was implemented.¹⁶ The signal would be expected at a frequency $f = v_{L,Si[110]}/p$ which
33 is 45.5 GHz.

34 Comparison of the measured frequencies of Table II with simulated frequencies using the
35 CGMD model allows us to label each reported frequency with a particular surface mode type
36 from the simulation. The superscripts and subscripts identifying each SAW frequency’s
37 wavelength and mode type are explained in the caption of Table II. While RL-2 (Sezawa) waves
38 (superscript *b* in Table II) in this frequency range have previously been observed by Brillouin

1 **Figure 5**



2

3 Figure 5: (a) ΔR for the $p/d = 200$ nm/60 nm sample using 400 nm probe in the S-Polarized and
4 P-polarized configurations. (b) Fourier transform amplitude of the signals in (a).

5

6

7

8

9

1 **Table II.**

Sample (p/d nm)	400 nm P-pol. (GHz)	400 nm S-pol. (GHz)	800 nm P-pol. (GHz)	800 nm S-pol. (GHz)
1000/60	^{1a} 4.5 , 8.6 ^{2a}	^{1a} 4.5 , ^{2a} _{4a} 8.1 , 12.2 , ^{3a} 15.2	^{1a} 4.7 , ^{2a} _{3a} 8.2 , 12.3	^{1a} 4.7 , ^{2a} _{3a} ^{4a} 8.2 , 12.1 , 20.2
600/60	^{1a} 7.6	^{1a} 7.6 , ^{2a} 13.7 , ^{3a} 18.2	^{1a} 7.7 , ^{2a} _{3a} 12.7 , ^{4a} 18.4 , 27.8	^{1a} 7.7 , ^{2a} _{3a} ^{4a} 13.3 , 18.5 , 27.8
400/112	^{1a} 9.8 , ^{2a} 16.4	^{1a} 10.0 , ^{2a} _{3a} 16.4 , 22.8	^{1a} 10.1 , ^{2a} 16.4	^{1a} 10.1 , ^{2a} 16.4
300/112	^{1a} 12.3 , ^{2a} 21.1	^{1a} 12.3 , ^{2a} 21.1 , ^{2b} 32.9	^{1a} 12.4 , ^{2a} 21.7	^{1a} 12.4 , ^{2a} 21.8
200/112	^{1a} 17.7	- , ^{2a} 30.0	^{1a} 17.7	^{1a} 17.1 , ^{2a} 30.4
200/60	^{1a} 18.8	^{1a} 18.8 , ^B _{1b} 26.4 , ^{2a} 29.9 , ^B _U 32.5 , ^{2b} 41.6 , ^B 48.8 , 55.4	^{1a} 18.8	^{1a} 19.1 , ^B _{2a} 26.7 , ^{1b} 30.5 , _U ^{2b} 32.5 , ^B 44.9 , 49.3 , 55.6
180/112	^{1a} 19.7 , ^{2a} - , ^{2b} 37.9 , 47.9	- , ^{2a} - , 35.3	^{1a} 19.5 , ^{2a} _{2b} - , 36.1 , 49.5	^{1a} 19.0 , ^{1b} _{2b} 26.7 , ^{2a} 35.4 , 49.0
170/112	^{1a} 20.8 , ^{2a} 38.0	- , ^{2a} - , 38.0	^{1a} 20.5 , ^{2a} 38.1	^{1a} 19.8 , ^{2a} 38.7
160/112	^{1a} 21.8	- , ^B - , 36.3 , ^{2a} 39.4	^{1a} 21.5 , ^B _{2a} - , 36.0 , ^{2b} 37.7 , 49.1 ,	^{1a} 21.0 , ^{1b} _{2a} 29.4 , ^{2b} 35.7 , 38.9 , 48.4
140/112	^{1a} 23.9	- , ^B - , - , 39.4	^{1a} 23.6	^{1a} 24.1 , ^{1b} _{2a} 32.6 , ^B 38.9 , 44.1

2

3 Table II: All measured frequencies from the four configurations of the experiment. A “-“
 4 indicates that a lower frequency that was present in another configuration is missing from that
 5 configuration. Superscript key: 1: $\lambda = p$, 2: $\lambda = p/2$, 3: $\lambda = p/3$, 4: $\lambda = p/4$, a = RL-1 mode, b = RL-
 6 2 (Sezawa) mode, B = independent bar mode, U = unidentified mode

1 scattering,^{17,18} the direct detection of coherent Sezawa waves above 10 GHz is not common. A
2 22.4 GHz signal from only one of the samples studied in Ref. 6 was found to agree with
3 predictions from a highly simplified simulation of a Sezawa mode of that nanostructure. Previous
4 studies using a transient grating technique more definitively detected Sezawa waves at
5 frequencies close to 1 GHz.^{19,20} As Table II indicates, however, in the present experiment we
6 have identified at least one Sezawa mode on five of the ten samples. These modes range in
7 frequency from 30 GHz to 48 GHz and are found to match the CGMD simulated frequencies to
8 better than 10% in all cases. Figure 3 includes the eight detected RL-2 (Sezawa) modes
9 alongside the phase velocity graph for that mode for the isotropic elastic calculation for SiO₂ on
10 Si, and the trend is comparable.

11 The data on three of the samples ($p/d = 200$ nm/60 nm, 160 nm/112 nm, and 140 nm/112 nm)
12 exhibit frequencies that do not correspond to surface wave-like motion, but rather to modes of an
13 individual bar attached to an SiO₂/Si substrate. These frequencies are marked with the
14 superscript *B* in Table II. In Ref.1, Lin et. al. determined that the vibrational frequencies of gold
15 bars on SiO₂ that they detected were not collective oscillations of many bars, but were instead
16 either flexural (F) or longitudinal (L) modes of individual free bars perturbed by contact with the
17 substrate. Figure 6 of Ref.1 shows the displacement patterns of the F and L modes, and Fig. 7 of
18 Ref. 1 shows how those mode frequencies vary as a function of the Young's modulus of the
19 substrate. To justify our assertion that these particular frequencies correspond to these individual
20 bar modes, we have run our CGMD simulation for an isolated bar in order to remove the
21 possibility of confusion with the collective SAW modes. In Table III, we compare the simulated
22 frequencies for the case of isolated and non-isolated bars and find excellent agreement between
23 the two versions of the simulation for these modes. The fact that the measured frequencies are
24 15% larger than the simulated ones for the $p/d = 160$ nm/112 nm and 140 nm/112 nm samples
25 may indicate a systematic error in the reported width of those bars.

26 While our ability to detect particular modes does seem to have a sample to sample variation that
27 prevents us from making definitive statements about the conditions for mode detection, one
28 obvious trend can be observed. For both wavelengths, the S-polarized probe is more sensitive to
29 higher frequencies than the P-polarized probe. In fact, at 400 nm, there are five cases for which
30 the S-polarized probe shows no (or barely distinguishable above the noise level) sensitivity to the
31 RL-1 SAW with $\lambda = p$, in spite of sensitivity to one or more higher frequency modes and despite
32 there being a strong signal with P-polarization. Figure 6 illustrates this effect for the $p/d = 180$
33 nm/112 nm sample; the P-polarized signal is dominated by the 19.7 GHz signal (due to an RL-1
34 SAW with $\lambda = 180$ nm), and is accompanied by 2 higher frequency modes, but the S-polarized
35 exclusively carries the 35.3 GHz signal (due to an RL-1 SAW with $\lambda = 90$ nm).

36 Polarization effects in picosecond ultrasonic experiments on nanostructures have been discussed
37 before,^{3,13,21,22} but a thorough explanation remains absent from the literature. The ΔR signal

1 **Table III.**

Sample (p/d nm)	Al Bar Cross section (nm)	CGMD Sim Freq. (GHz)	CGMD Sim Freq. Isolated (GHz)	Meas. Freq at 400 nm probe (GHz)
200/60 F	100 × 25	27.8	26.9	26.4
200/60 L	100 × 25	55.2	54.9	55.4
160/112 F	80 × 25	32.4	31.9	36.3
140/112 F	70 × 25	36.2	35.9	39.4

2

3 Table III: Individual bar mode summary for the four modes labeled with the superscript B in
4 Table II.

5

6

7

8

9

10

11

12

13

14

15

16

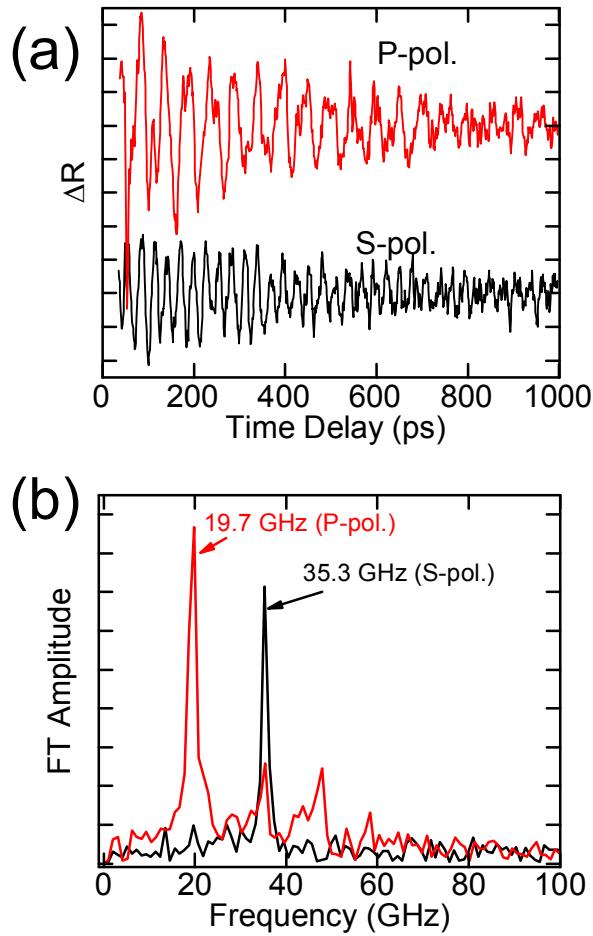
17

18

19

20

1 **Figure 6**



2

3 Figure 6: (a) ΔR for the $p/d = 180 \text{ nm}/112\text{nm}$ sample using 400 nm probe in the S-Polarized and
4 P-polarized configurations. (b) Fourier transform amplitude of the signals in (a).

5

6

7

8

9

10

1 caused by the SAW should have two primary components – a piezo-optic component caused by
2 strain induced changes in the optical constants of the Al lines, and a displacement component
3 caused by the vertical and lateral movement of the lines themselves during the surface wave
4 motion. The piezo-optic component is expected to be much stronger for an 800 nm probe rather
5 than a 400 nm probe due to the d-band transition in Al near 1.5 eV^{23,24} and this is the reason why
6 the 800 nm data shows no missing low-frequencies in column 4 of Table II, unlike the 400 nm
7 data in column 2.

8 The strong effect of probe polarization on the experiment is not surprising when we consider the
9 shape of the electric and magnetic field functions in the near field of the optical grating presented
10 by this relatively simple nanostructure. While an accurate prediction of ΔR as a function of the
11 time-dependent strain caused by the SAWs is beyond the scope of this work, we have performed
12 finite element calculations using COMSOL of the probe laser field amplitude for the samples in
13 the experiment. Figure 7 shows the normalized field amplitude for S-polarized and P-polarized
14 400 nm light incident on the $p/d = 180 \text{ nm}/112 \text{ nm}$ sample. In the P-polarized case, the field near
15 the lines takes on a periodicity similar to that of the lines, while in the S-polarized case there are
16 sharp peaks in the field amplitude near the edges of the Al lines. These differences in the field
17 near the Al lines for the S and P cases when coupled with the oscillatory motion of the lines due
18 to the presence of a SAW are expected to cause changes in our far field sensitivity to SAWs with
19 differing wavelengths and displacement patterns. In certain cases, such as the one illustrated in
20 Fig. 6, these differences appear to completely reduce or eliminate our far-field sensitivity to the
21 lowest frequency SAW available to the structure.

22 **Summary**

23 In summary, we have used picosecond laser ultrasonics to generate and detect high frequency
24 SAWs in the GHz range on a patterned layered nanostructure of Al/SiO₂/Si. We have detected
25 Rayleigh-like and Sezawa-like modes with frequencies as high as 50 GHz, as well as a number
26 of independent bar modes. We have compared our results to analytical calculations of an SiO₂/Si
27 structure and to coarse-grained molecular dynamics simulations of the complete structure in
28 order to accurately label the detected modes. We also found a strong dependence of the
29 sensitivity to specific modes on the polarization of the probe light, especially when using a
30 frequency-doubled probe at 400 nm.

31 **Acknowledgements**

32 The authors acknowledge the support of National Science Foundation award number DMR-
33 1709521 “RUI: Acoustic Phonons in Nanostructures: Surface Waves, Thermal Transport, and
34 Imaging.” The sample fabrication was performed at the Cornell NanoScale Facility, a member
35 of the National Nanotechnology Coordinated Infrastructure (NNCI), which is supported by the

1 National Science Foundation (Grant ECCS-1542081). We also acknowledge technical
2 assistance from Mr. Sushant Mahat.

3

4

5

6

7

8

9

10

11

12

13

14

15

16

17

18

19

20

21

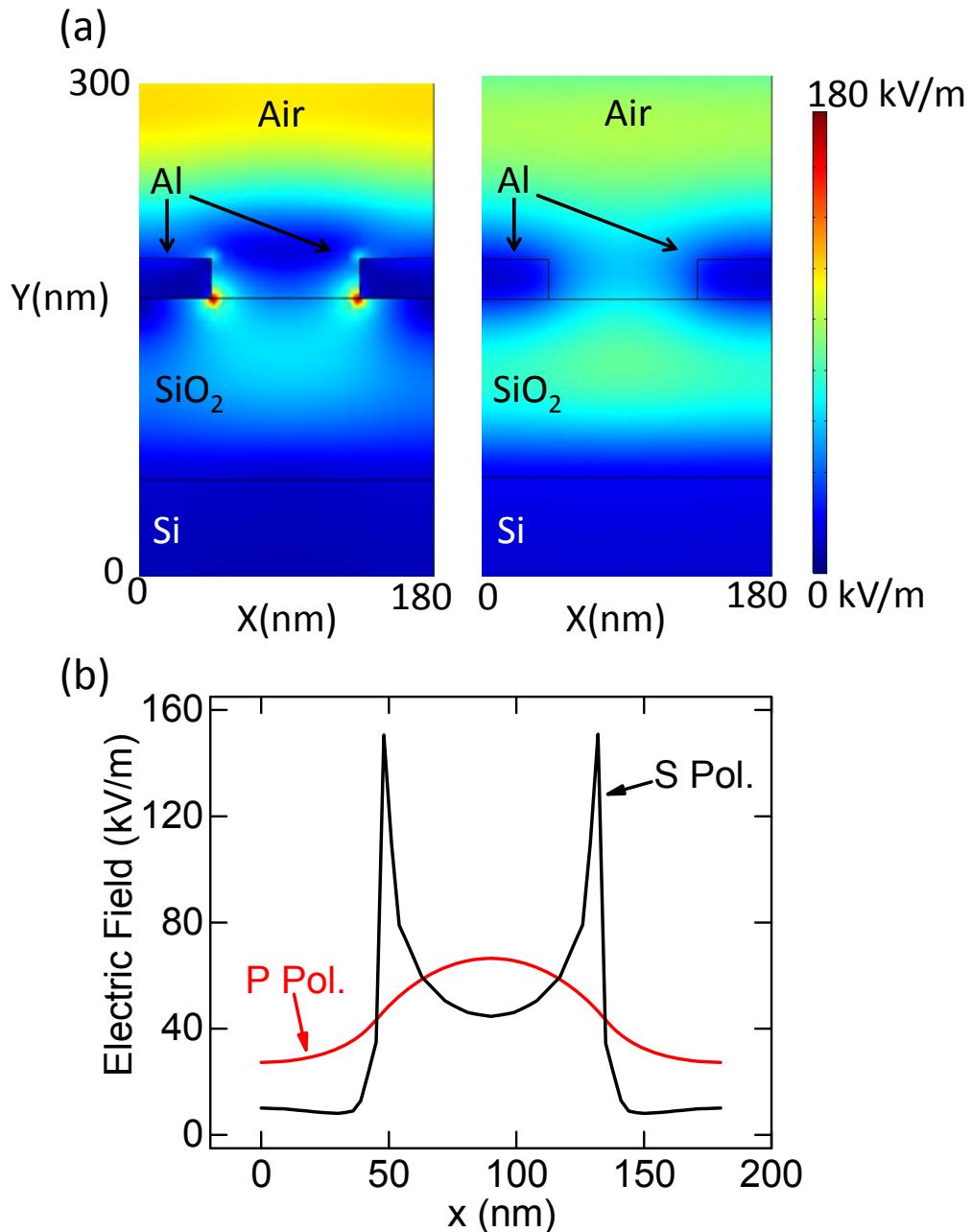
22

23

24

25

1 **Figure 7**



2

3 Figure 7: Finite element model of 400 nm probe light incident on $p/d = 180 \text{ nm}/112 \text{ nm}$ sample.
4 (a) Colormap of normalized electric field amplitude for S polarized (Left) and P polarized
5 (Right) probe beam configuration. (b) Electric field amplitude as a function of lateral position
6 taken at the Al pattern/SiO₂ interface for S and P polarizations.

7

¹H.-N. Lin, H. J. Maris, L. B. Freund, K. Y. Lee, H. Luhn, and D. P. Kern, *J. Appl. Phys.* 73, 37 (1993).

² B. Bonello, A. Ajinou, V. Richard, P. Djemia, and S. M. Cherif, *J. Acoust. Soc. Am.* 110, 1943 (2001).

³D. H. Hurley and K. L. Telschow, *Phys. Rev. B* 66, 153301 (2002).

⁴ J. Sadhu, J. H. Lee, and S. Sinha, *Appl. Phys. Lett.* 97, 133106 (2010).

⁵ M. E. Siemens, Q. Li, M. M. Murnane, H. C. Kapteyn, R. Yang, E. H. Anderson, and K. A. Nelson, *Appl. Phys. Lett.* 94, 093103 (2009).

⁶ M.M. Bjornsson et. al., *J. Appl. Phys.* 117, 095305 (2015).

⁷ B. A. Auld, *Acoustic Fields and Waves in Solids* (Krieger Publishing Company, Malabar, Florida, 1990), Vol. 2, Ch. 10.

⁸ G. W. Farnell and E. L. Adler, in *Physical Acoustics*, edited by W. P. Mason and R. N. Thurston (Academic Press, New York, 1972), Vol. 9, p. 35.

⁹ G. A. Antonelli, B. Perrin, B. C. Daly, and D. G. Cahill, *MRS Bull.* 31, 607 (2006).

¹⁰ C. Thomsen, H. T. Grahn, H. J. Maris, and J. Tauc, *Phys. Rev. B* 34, 4129 (1986).

¹¹ L.M. Brekovskikh, *Sov. Phys. Acoust.* 5, 288 (1960).

¹² D. Gelda, J. Sadhu, M.G. Ghossoub, E. Ertekin, and S. Sinha, *J. Appl. Phys.* 119, 164301 (2016).

¹³ G. A. Antonelli, H. J. Maris, S. G. Malhotra, and J. M. E. Harper, *J. Appl. Phys.* 91, 3261 (2002).

¹⁴ A. A. Maznev and A. G. Every, *J. Appl. Phys.* 106, 113531 (2009).

¹⁵ H. Robinson, Y. Hahn, and J.N. Gau, *J. Appl. Phys.* 65, 4573 (1989).

¹⁶ Q. Miao, L.-W. Liu, T.J. Grimsley, A.V. Nurmikko, and H.J. Maris, *Ultrasonics* 56, 141 (2015).

¹⁷ N.L. Rowell and G.I. Stegeman, *Phys. Rev. Lett.* 41, 970 (1978).

¹⁸ J.G. Dil, *Rep. Prog. Phys.* 45, 285 (1982).

¹⁹ A. A. Maznev, *Phys. Rev. B* 78, 155323 (2008).

²⁰ A. A. Maznev and O. B. Wright, *J. Appl. Phys.* 105, 123530 (2009).

²¹ D. Li, P. Zhao, J.-C. Zhao, and D. G. Cahill, *J. Appl. Phys.* 114, 143102 (2013).

²² Q. Miao, L.-W. Liu, T.J. Grimsley, A.V. Nurmikko, and H.J. Maris, *Ultrasonics* 56, 141 (2015).

²³ A. Devos, *Ultrasonics* 56, 90 (2015).

²⁴ A. Devos and A. Le Louarn, Phys. Rev. B 68, 045405 (2003).