Attenuation of Picosecond Ultrasonic Pulses in a thin Silicon Wafer

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Abstract
We report picosecond ultrasonic measurements of phonon attenuation in a 56 µm thick Si wafer coated with a 25 nm Al film. The measurements are of interest for picosecond ultrasonic imaging schemes, and for investigations of nanoscale heat transport. An ultrafast laser was used to perform the pump-probe experiment. A probe delay stage with a range of several nanoseconds allowed detection of coherent acoustic phonon (picosecond ultrasonic) pulses that had travelled one or two round trips through the Si. By comparing the signals from the first phonon pulse to the second phonon pulse we measured the attenuation of longitudinal acoustic phonons as a function of frequency (30GHz-90GHz) and temperature (100K-300K). We find that the frequency dependence at 100K is reduced compared with the frequency dependence at higher temperatures.

Keywords: Picosecond ultrasonics, ultrafast optics, pump-probe measurements, coherent acoustic phonons, phonon attenuation, silicon

1. Introduction and Background

The attenuation of high-frequency ultrasound in dielectric or semiconductor crystals is a topic with a considerable history that has relevance to two areas of current interest in applied solid state physics. First, long wavelength acoustic phonons with frequencies near 1 THz are expected to play an important role in nanoscale heat transport [1]. Second, the feasibility of imaging buried nanostructures by using high frequency acoustic waves will depend on attenuation levels at different temperatures and frequencies [2]. In this paper, we report picosecond ultrasonic measurements of attenuation in a thin, undoped Si wafer in the temperature range 100 K-300 K and frequency range 30 GHz – 90 GHz. Attenuation of acoustic phonons in this frequency range in Si has been studied previously by Hao and Maris, but only at temperatures lower than 130 K [3]. The highest frequency for which the attenuation in Si has previously been measured at room temperature is 20 GHz [4].

In a crystalline semiconductor such as Si, the dominant mechanism for the decay of an ultrasonic wave is expected to be its interaction with thermal phonons. A comprehensive review of the theory of attenuation of sound waves by thermal phonons has been written by Maris [5]. For ultrasonic waves at high frequencies and low temperatures such that the acoustic wave frequency \( \omega \approx kT/\hbar \), the attenuation is best described by three-phonon anharmonic decay processes as originally put forth by Landau and Rumer [6]. For longer wavelength ultrasound the decay mechanism is better described by a relaxation damping theory due to Akheiser [7], where one considers that the sound waves disturb the thermal phonons whose frequencies depend on strain. The thermal phonons then collide with one another, returning the system to equilibrium as energy is removed from the sound wave. For our measurements, we can assume that all...
ultrasound waves of interest are of a much lower frequency than the average thermal phonon frequency \((kT/h = 2.08 \text{ THz at } 100\text{K})\). This places us in a regime where both theories could be expected to accurately describe the attenuation.

Following Ref. 5, we can further subcategorize our frequency and temperature range into the regimes \(\omega\tau >> 1\) and \(\omega\tau << 1\) where \(\tau\) is the average lifetime of the thermal phonons in the crystal. Using calculations based on thermal conductivity measurements of Si, we use the expression \(\tau^{-1} = B\omega_h^2T\) to determine \(\tau\) [8]. We choose the value of \(B\) given for normal three-phonon processes which is \(2.4\times10^{-19} \text{ s K}^{-1}\). This results in a range of \(\tau = 9.7\times10^{-12} \text{ s at } 300\text{K}\) to \(\tau = 2.6\times10^{-10} \text{ s at } 100\text{K}\). We then have a minimum value of \(\omega\tau = 1.83\) for 30 GHz ultrasound at 300 K, and a maximum value of \(\omega\tau = 147\) for 90 GHz ultrasound at 100K. Thus, for the most part, we find that the assumption of \(\omega\tau >> 1\) will hold for our experiment, but we note that for the more elevated temperature measurements, which are of particular interest for many applications, we find ourselves in an intermediate regime.

Ultrasonic attenuation in Si due to anharmonic three phonon processes should have a quadratic dependence on frequency regardless of the frequency or temperature range. This is the result of an important paper by Conyers Herring [9] which extended the anharmonic phonon decay model of Landau and Rumer to account for the types of phonon collisions that are allowed and disallowed by the crystalline anisotropy. In contrast, ultrasonic attenuation in Si as described by Akhiezer’s relaxation damping should exhibit a quadratic dependence for \(\omega\tau << 1\) but should exhibit no frequency dependence for \(\omega\tau >> 1\). The full expression for the attenuation \(\alpha\) using relaxation damping theory is given in Eq. 130 of Ref. 5. Here we reproduce a more simplified expression that is valid for all values of \(\omega\tau\) when the group velocity of the thermal phonons is much less than the velocity at frequency \(\omega\). The attenuation \(\alpha\) is given by

\[
\alpha = \frac{CT}{2\rho s^3} \frac{\omega^2\tau}{1 + \omega^2\tau^2} (\langle \gamma^2 \rangle - \langle \gamma \rangle^2)
\]

where \(C\) is the volumetric heat capacity, \(\rho\) is the density, \(s\) is the speed of the ultrasound wave, and \(\gamma\) is the Grüneisen parameter [10]. The brackets \(\langle ... \rangle\) indicate averages taken over the entire thermal phonon spectrum and all 3 three polarizations. The term in front of the parentheses in Eq. 1 reduces to \(CT/2\rho s^3\tau\) for \(\omega\tau >> 1\) and to \(CT\omega^2\tau/2\rho s^3\) for \(\omega\tau << 1\).

2. Experiment

Our measurements were made using an ultrafast optical pump-probe experiment at the Frederick Seitz Materials Research Laboratory at the University of Illinois at Urbana-Champaign that has been described previously [11]. The sample was an undoped Si wafer that had been thinned to 56 µm and polished on both sides. The sample was coated with a 25 nm Al film to be used as a transducer. A mode-locked Ti:sapphire oscillator with a repetition rate of 80MHz was used to perform the pump-probe
experiment. Pump pulses from the oscillator were used to rapidly heat the Al film, generating longitudinal acoustic phonon pulses. These acoustic pulses have a roughly single-cycle character with a compressive component leading the rarefacting component, and the bandwidth of the pulse is ~100 GHz. The thickness of the Si was chosen so that phonon pulses completed a single round trip through the Si in a little more than the 12.5 ns repetition period of the oscillator. A probe delay stage with a range of a few nanoseconds allowed detection of phonon pulses that had travelled one (13.3 ns) or two (26.6 ns) round trips through the Si.

![Figure 1](image.png)

Figure 1. Pump-probe reflectivity data showing the acoustic phonon pulses after one (13.3 ns) and two (26.6 ns) round trips in the Si wafer. The signals at different temperatures are multiplied by the factors indicated on the graph for comparison.

Fig. 1 shows the reflectivity changes ($\Delta R(t)$) caused by the travelling phonon pulses after the first and second round trips at 5 temperatures ranging from 100 K to 300 K. $\Delta R(t)$ is a convolution of the amplitude of the travelling acoustic pulse and the optical sensitivity function of the Al [12]. The thermal backgrounds of the data have been removed and each data set has been offset for visual convenience.

3. Results and Discussion

A number of qualitative observations can be made upon inspection of Fig. 1. First, the strong temperature dependence of the attenuation is indicated by the multiplicative factors that have been placed on the figure. Second, the variation in arrival times as a function of temperature is indicative of the temperature dependence of the sound velocity. We find a roughly 0.5% change in the longitudinal sound velocity between 300 K and 100 K which is comparable a value obtainable by extrapolating the lower temperature data of Fig. 8 of Ref. 3. Lastly we note the slight asymmetry in the 1st echo that shifts as the temperature increases. The asymmetric echo shape is caused in part by the fact that the initial acoustic pulse is not a perfect single-cycle due to surface roughness and the presence of an oxide at the free surface. The second cause of asymmetry in the echo is the effect of acoustic dispersion, which causes the higher frequency components to trail the lower frequency components. The fact that the 300K signal appears larger at the early part of the echo while the 100K signal appears larger at
the later part of the echo indicates that the higher frequencies are preferentially attenuated at the higher temperatures.

To study the ultrasonic attenuation of Si [100] in more detail, we follow the method of Refs. 3 and 4 and analyze Fourier transforms \( \Delta R(\omega) \) of the data. Fig. 2 shows Fourier transform magnitudes for the first and second pulses at 100 K and 300 K. We determine the attenuation as a function of frequency by the formula

\[
\alpha(\omega) = \frac{1}{d} \ln \left( \frac{\Delta R_2(\omega)}{\Delta R_1(\omega)} \right),
\]

(2)

Where \( d \) is the round trip distance through the Si wafer (112 \( \mu \)m), and \( \Delta R_1(\omega) \) and \( \Delta R_2(\omega) \) are the magnitudes of the Fourier transforms of the first and second pulses. In using this expression for the attenuation we ignore any losses that occur at either the free surface or the Al film interface. In Fig. 3 we plot \( \alpha \) as a function of frequency for temperatures ranging from 100 K – 300 K. The overall attenuation at 100 K is somewhat larger than in Ref. 3 in which the authors reported a value of \( \alpha = 4 \) cm\(^{-1}\) for 50 GHz ultrasound at 90K and a value of \( \alpha = 9 \) cm\(^{-1}\) for 50 GHz ultrasound at 110 K as compared to the present value of 42 cm\(^{-1}\). It is likely that this discrepancy is in part attributable to the fact that both our analysis and that of Ref. 3 have ignored surface effects. This would have a more significant impact on the present data due to the 112 \( \mu \)m difference in propagation length between the first and second signal as compared with the 630 \( \mu \)m difference in propagation length of Ref. 3. The room temperature measurement shows better agreement with the measurement of Ref. 4, which gives a value of 80 cm\(^{-1}\) for 20 GHz at 300 K as compared with the present value of 102 cm\(^{-1}\) for 30 GHz at 300 K. We also note that \( \alpha \) increases quite linearly with \( T \) at 50 GHz, which is near the peak of the detected Fourier spectrum. This linearity is less clear at the lower and higher frequencies where the signal is not as large.

Figure 2. Fourier transforms of the first two \( \Delta R \) pulses for 100K and 300K.
Despite some scatter within the data, Fig. 3 indicates that the frequency dependence of the attenuation changes with temperature, especially as the temperature is reduced to 100 K. In fact, the frequency dependence is quite flat at 100K, which is in agreement with the 30 K – 130 K measurements of Ref. 3. Above this temperature, however, we observe an increasing attenuation with frequency. While our data are not conclusive as to whether a quadratic frequency dependence is demonstrated, it is still possible to make some conclusions on the nature of the acoustic attenuation mechanism. If the Herring process of anharmonic decay of the acoustic phonon pulse were responsible, a consistent quadratic dependence should be observed. This mechanism should display itself more clearly for larger values of $\omega \tau$ and so we would expect the effect to be easier to observe at 100 K rather than more difficult. On the other hand, the Akheiser mechanism of relaxation damping described by Eq. (1) does predict that there should be a transition from a frequency independent attenuation to a quadratic dependent attenuation as $\omega \tau$ decreases from greater than 1 to less than 1. Despite our earlier estimate that the lowest experimental value of $\omega \tau$ was 1.68, it seems very likely that this is the nature of the transition we have observed.

To support the conclusion that the Akheiser mechanism of relaxation damping is primarily responsible for our measured attenuation, we can attempt to use Eq. 1 to predict the overall magnitude of the attenuation. The Gruneisen parameters for Si vary with mode and frequency from slightly less than -1 to slightly more than +1. If we crudely assume that the factor in parenthesis in Eq. 1 is simply equal to 1 we find that it predicts a value of 143 cm$^{-1}$ for 30 GHz ultrasound at 300 K. This overestimates the measured attenuation by less than a factor of two, which is reasonable given our speculative treatment of the Gruneisen parameter term. The attenuation based on the Herring process in Si is predicted to be two orders of magnitude lower than the measured value [3,10].
4. Conclusion

In summary, we have used picosecond ultrasonics to measure the attenuation of longitudinal acoustic phonons in Si from 100 K up to 300 K. The frequency dependence of the attenuation is found to vary with temperature in a manner that is consistent with relaxation damping.

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