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Bolometric detection of picosecond acoustic pulses in silicon and gallium arsenide

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Abstract. In this paper, we show that superconducting Aluminium bolometers are effective detectors of picosecond acoustic pulses. Femtosecond pulsed optical excitation of a thin Chromium film deposited on a crystal (Si or GaAs) substrate results in the generation of ballistic phonon pulses. Bolometers fabricated on the face of the substrate opposite the Cr film were used to detect the pulses. The bolometer signal was seen to consist of two components: a conventional heat pulse and, arriving at the bolometer slightly earlier, a “fast” longitudinal polarized pulse. Phonon imaging measurement showed the fast pulse to be highly collimated over propagation distances exceeding 1 cm. We attribute this fast signal to the picosecond acoustic pulse generated by the thermoelectic effect in the Cr transducer. We speculate as to the possible physical mechanisms of its detection by the bolometer.

1. Introduction
Acoustic pulses of picosecond duration can be generated by ultrafast heating of a thin metal film transducer using femtosecond laser pump pulses [1]. The resulting rapid thermal expansion of the film launches a strain pulse into the underlying substrate material. The conventional method of detecting picosecond acoustic pulses is to use a femtosecond optical probe pulse. The strain pulse modulates the probe beam reflectance which is measured as a function of the pump to probe delay time [1-3]. Typically, picosecond acoustic pulses have frequency components up to about 100 GHz and may be used to study the acoustic properties of materials [4] and also probing nanostructures [5]. The pump-probe technique has the main advantage of very high temporal resolution, but can be difficult to implement for long propagation distances due to the large

Figure 1. Bolometer signals due to ultrafast excitation of Cr film on c-axis sapphire for different values of the pump energy density absorbed in the Cr film [9], the propagation distance in this case was 10 mm. The LA heat pulse and “fast” pulse are indicated.
optical delay lines required and the need for precise optical alignment. Alternative methods for detecting ultrafast acoustic pulses include Brillouin scattering [6] and luminescence techniques [7].

Superconducting bolometers have long been the detector of choice for use in heat pulse spectroscopy [8]. Their primary advantages being: simplicity, high sensitivity and, since they can be fabricated with micron dimensions, high spatial resolution. The response time of bolometers, though entirely satisfactory for nanosecond heat pulse measurements, is considered far too slow for use in picosecond acoustics. However, recently we showed that superconducting Aluminium bolometers with sub-nanosecond rise time could detect picosecond acoustic pulses propagating in Sapphire [9]. Figure 1 shows the bolometer-detected acoustic signal resulting from femtosecond pulsed laser excitation of a Chromium film after propagation through 1 cm of c-axis Sapphire. The narrow pulse arriving a few ns ahead of the ballistic heat pulse was attributed to the coherent picosecond acoustic pulse.

In this work we use superconducting bolometers as detectors to investigate the propagation of picosecond acoustic pulses in Si and GaAs. We also consider the possible mechanisms of detection.

2. Experimental details
The experimental setup, shown schematically in figure 2, is basically the same as a conventional heat pulse experiment using a laser heated metal film as the phonon source. However, instead of using a ns-duration pulsed laser, a femtosecond laser was used to excite the metal film.

The Silicon sample used in this work was a (001)-oriented, 5 mm-thick crystal, lightly p-doped with resistivity > 1000 Ω cm. Opposite (001) faces were polished parallel to each other and after polishing the thickness of the crystal was measured using a micrometer to be 4.65 mm. On one polished surface was deposited by vacuum evaporation a 180 nm-thick film of Chromium. On the opposite face was deposited a granular Aluminium bolometer with an active area of 0.5 mm × 0.05 mm. The Aluminium was evaporated from an Alumina crucible source and was 30 nm-thick giving a normal state resistance of about 200 Ω. The (001)-oriented semi-insulating Gallium Arsenide crystal was also 5 mm-thick before processing. It was prepared and the Cr transducers and Al bolometers were fabricated using the same procedure as for the Si sample, described above. After polishing, the crystal was 4.48 mm in thickness.

For the measurements, the samples were mounted in an optical-access cryostat and held at a constant temperature of about 2.2 K which was on the superconducting transition-edge of the bolometer where its DC resistance was about 50 Ω. The bolometer was DC biased with a current of about 10 μA. An amplified Ti:Sapphire laser (Coherent Libra®) was used for optically pumping the Cr film. The laser produced 120 fs, λ = 800 nm pulses of 1 mJ energy and repetition rate 1 kHz. The pulses were attenuated using neutral density filters, passed through a 1 mm-diameter aperture and then focussed to a spot of diameter ~ 200 μm on the Cr film. Using this arrangement it was possible to achieve an absorbed energy density up to about 2 mJ cm⁻², above which the Cr film was damaged. The position of the excitation spot on the film was set using galvanometer x-y scanning mirrors, which enabled phonon imaging measurements to be made.

The bolometer signal, which was a transient voltage pulse, was amplified and processed using a 2 GHz-bandwidth pre-amplifier and a 0.5 GS/s digitizer, triggered from the laser pulse using a photodiode.
3. Results

Figure 3 shows typical bolometer signals as a function of time for the Si sample at two different values of the absorbed energy density, 0.009 and 0.028 mJ cm\(^{-2}\). These were obtained with the excitation spot positioned exactly opposite the active part of the bolometer. At the lower absorbed energy density, two pulses are seen. These are the longitudinal acoustic (LA) and, much stronger, transverse acoustic (TA) heat pulse signals propagating along [001], and are due to heating of the Cr film by the laser pulse. The times of the start of the rising edge of the LA and TA heat pulses are respectively \(\tau_{\text{LA}} = 550 (\pm 1)\) ns and \(\tau_{\text{TA}} = 784 (\pm 1)\) ns, which correspond roughly to the LA and TA sound velocities in Si. At the higher absorbed energy density, the TA signal saturates the bolometer, and a third, very narrow, “fast” pulse is observed arriving about 8 ns earlier in time than the LA heat pulse. This is shown more clearly in the inset to figure 3 which zooms in close to the LA time of flight. The time of flight of the fast pulse is \(\tau_{\text{F}} = 542 (\pm 1)\) ns and the rise and fall times are similar to those of the breakthrough of the femtosecond optical pulse seen at \(t = 0\), so would appear to be limited by the response time of the bolometer and its associated signal processing electronics. On the decaying tail of the TA pulse, sharp peaks are seen at 1633 (±1) ns and 2723 (±1) ns, see figure 4, due to reflections of the fast pulse at the surfaces of the crystal.

![Figure 3](image3.jpg)

Figure 3. Bolometer signals (offset for clarity) after acoustic pulse propagation through a (001) Si wafer of thickness 4.65 mm at absorbed laser energy densities of 0.009 mJ cm\(^{-2}\) (lower trace) and 0.028 mJ cm\(^{-2}\) (upper trace). The start of the LA and TA heat pulse.

![Figure 4](image4.jpg)

Figure 4. Reflections of the fast pulse after 3 crossings of the Si wafer (1st) and 5 crossings of the wafer (2nd).

Figure 5 shows the peak amplitudes of the fast pulse and LA heat pulse as a function of the excitation energy density. It is clear that the fast pulse appears only above a threshold in the absorbed
excitation energy density, in this case about 0.02 mJ cm\(^{-2}\). The value of the threshold was found to depend on the laser pulse width: increasing the pulse width to 4 ps by detuning the compressor in the amplifier resulted in a small increase of the threshold. The spatial distribution of the phonon flux in the fast pulse was measured using the phonon imaging technique [10] and is shown in figure 6. The fast pulse was only detected when the excitation spot was directly opposite the active part of the bolometer, which means that it is highly directional and propagates normal to the Cr transducer.

In the case of the GaAs crystal, the fast pulse was weaker and much less well resolved, but, at high pump energy density, can be clearly seen about 5 ns ahead of the leading edge of the LA heat pulse, figure 7. Interestingly, there was no clear evidence of a threshold in the pump fluence for observation of the fast pulse in GaAs (inset to figure 7), and, unlike for Si, the fast pulse never became stronger than the LA heat pulse, even at the highest pump fluence. Due to the poor resolution of the fast pulse it was not possible to make an image. However, it was clear that, as in the case of Sapphire and Si, the fast pulse was only detected when the excitation spot was directly opposite the active part of the bolometer.

**Figure 5.** Dependence of the amplitudes of the LA heat pulse and the fast pulse in Si as a function of the absorbed pump energy density. The LA pulse amplitude has been multiplied by 50 times to put it on the same scale as the fast pulse. The threshold for the appearance of the fast pulse is observed at an energy density of about 0.02 mJ cm\(^{-2}\).

**Figure 6.** The amplitudes of the LA heat pulse and fast pulse in Si as a function of the position of the pump spot. The scan is taken along the x-direction, through the centre of the bolometer (along the horizontal as shown in figure 2). The position and size of the bolometer are indicated by the hatched area. Notice how the fast pulse is only detected when the pump spot is directly opposite the bolometer.

**Figure 7.** Close-up of the LA bolometer signal after propagation through a (001) GaAs crystal of thickness 4.48 mm. The fast pulse is barely resolved on the leading edge of the LA heat pulse. The inset shows the amplitude of the fast pulse as a function of the absorbed energy density, no threshold is observed.
4. Discussion

We now consider these results with the aim of reaching conclusions as to the possible nature of the fast pulse and the mechanism of its detection by the bolometer. In our earlier work using a Sapphire crystal [9], the fast pulse was attributed to the coherent picosecond acoustic pulse generated by the photoelastic effect in the Cr film. The imaging measurements are certainly consistent with this also being the case for the Si and GaAs samples discussed here. The fast pulse is only detected when the excitation spot is directly opposite the bolometer, which means, unlike for the heat pulse, the phonon wavevectors are directed normal to the plane of the film. Since the excitation spot is much larger than the typical phonon wavelength and the acoustic pulse is spatially coherent across the excitation front, there is no significant spreading of the pulse due to diffraction effects.

Mechanisms for the detection of the fast pulse by the bolometer are, however, not immediately obvious. The maximum frequency of the Fourier components of a picosecond acoustic pulse depends on such things as the metal film thickness, the optical penetration in the film and the speed of thermal diffusion in the metal [11]. Typically, the maximum frequency is in the region of 100 GHz, which is less than the superconducting gap of the bolometer material, \( \Delta / \hbar \approx (3k_B T_c / \hbar)(1 – T / T_c)^{1/2} \approx 180 \text{ GHz} \).

For comparison, to generate a heat pulse having a frequency at the peak of the Planck distribution of 100 GHz would require a heater temperature of 1.6 K, which is less than the sample temperature in this experiment. Hence absorption of the picosecond pulse in the bolometer is not expected to result in a stronger signal than for the heat pulse as is observed in the case of Si. It is possible that at very high strain amplitude nonlinear acoustic effects in the bolometer give rise to the detection. This would be consistent with the threshold. Another possibility is that the strain could be directly influencing the superconducting transition or critical current of the Al bolometer. An alternative explanation is that acoustic nonlinearity in the crystals gives rise to the formation, at high strain amplitude, of an acoustic packet with strain solitons at the leading edge followed by an oscillatory tail [12, 13, 6, 7].

It has been shown that the acoustic solitons can contain phonon frequency components up to about a THz [7], which the bolometer is easily capable of detecting. We can arrive at a conclusion as to whether the fast pulse is due to solitons or a normal picosecond ultrasonic pulse by careful consideration of the velocities.

For the Si sample, the time of the start of the rise of the LA heat pulse corresponds to a sound velocity of 8450 (±25) ms\(^{-1}\), which is a little less than the speed of longitudinal sound along [001] Si, 8486 ms\(^{-1}\). The latter was calculated using the accepted values of the density, \( \rho = 2329 \text{ kg m}^{-3} \), and the low temperature elastic constant, \( C_{11} = 1.677 \times 10^{11} \text{ J m}^{-3} \), obtained from recent low-frequency acoustic velocity measurements [14]. The slightly smaller velocity for the heat pulse is probably due to the effects of acoustic anisotropy and also scattering of the high frequency phonons in the heat pulse. The measured velocity of the fast pulse is 8580 (±25) ms\(^{-1}\), which is about 1% higher than the calculated value. This provides strong evidence that we are detecting an acoustic packet containing acoustic solitons at its leading edge. The solitons, which form within a very short distance, \( \sim 100 \mu \text{m} \), at our pump energy density, propagate at a speed that is higher than the speed of low-frequency longitudinal sound, \( c_{LA} \). For [001] propagation in a cubic crystal, the soliton speed is given by [13]

\[
u = c_{LA} + \frac{\eta_0 (3C_{11} + C_{111})}{6\rho c_{LA}},
\]

where \( \eta_0 \) is the initial strain amplitude and the third-order elastic constant, \( C_{111} = -4.22 \times 10^{12} \text{ kg m}^{-1}\text{s}^{-2} \). A 1% increase in velocity compared to \( c_s \) suggests that the peak strain amplitude \( \eta_0 \sim 0.25\% \), a figure typical of that obtained using an amplified Ti:Sapphire laser to excite the Cr transducer [6, 7]. Increasing the pump energy density further above the threshold does not lead to the soliton travelling still faster, but the formation of additional solitons (a soliton train). Further evidence for the soliton interpretation is provided by consideration of what happens when the acoustic packet is reflected at the sample boundaries. The strain pulse reverses phase on reflection from the sample/vacuum interface. Therefore on reflection of the pulse from the bolometer side, the phase reversal results in destruction of the solitons and the acoustic packet returns at the normal speed of sound. When the pulse is
reflected once more, this time at the Cr film side of the sample, the phase is again reversed. Therefore the solitons can form again and the acoustic packet returns to the bolometer at the higher speed. The measured arrival times of the first reflected pulse $1633\,\text{ns} = 2\tau_F + \tau_{LA}$ and, for the second reflected pulse, $2723\,\text{ns} = 3\tau_F + 2\tau_{LA}$ are fully consistent with these considerations.

For the GaAs sample, the velocity of the fast pulse is $4780\,\text{(±25) ms}^{-1}$, very close to the velocity of the picosecond acoustic pulse measured in reference [4]. The LA heat pulse velocity was measured as $4740\,\text{(±25) ms}^{-1}$. As is the case of Si, this is slightly less than the velocity of low-frequency sound. Therefore, in GaAs, it appears the fast pulse is due to the coherent picosecond acoustic pulse, which, with its frequency components predominantly below 100 GHz, propagates at a speed close to that obtained in low-frequency ultrasonic measurements. A possible reason for not observing the solitons in GaAs is that, due to the strong scattering of THz phonons in GaAs, the soliton decays. The mean free path of 650 GHz phonons in GaAs is only 0.8 mm [15]. Therefore, assuming the mean free path $\sim \nu^{-4}$ (isotope scattering), the maximum frequency that can propagate through the sample is about 400 GHz. This interpretation is also consistent with there being no clear pump threshold for observation of the fast pulse in GaAs. The weak response of the bolometer to the pulse is probably due to there being a small amount of spectral content of the picosecond acoustic pulse in 150-200 GHz range as has been observed in pump-probe experiments [5].

5. Conclusion
We have shown that superconducting Al bolometers can be used to detect picosecond ultrasonic pulses propagating in Si and GaAs. Therefore bolometers provide a detection method that is complementary to the established techniques such as optical pump-probe techniques. In the case of Si, the high amplitude strain pulses evolve into an acoustic packet consisting of solitons at the leading edge, which propagate slightly faster than the speed of low-frequency longitudinal-polarized sound. The high-frequency acoustic components in the solitons are efficiently detected by the bolometer. In the GaAs sample, the strong phonon scattering results in attenuation of acoustic components above 400 GHz and the soliton is destroyed. However, the bolometer still responds weakly to the picosecond acoustic pulse.

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References
[1] Thomsen C, Grahn H T, Maris H J and Tauc J 1986 Phys. Rev. B34 4129.
[2] Tas G and Maris H J 1994 Phys. Rev. B49 1056.
[3] Saito T, Matsuda O and Wright O B 2003 Phys. Rev. B67 205421.
[4] Hao H –Y and Maris H J 2001 Phys. Rev. B63 224301.
[5] Huynh A, Lanzillotti-Kimura N D, Jusserand B, Perrin B, Fainstein A, Pascual-Winter M F, Peronne E and Lemaitre A 2006 Phys. Rev. Lett. 97 115502.
[6] Muskens O L and Dijkhuis J 2004 Phys. Rev. B70 104301.
[7] Muskens O L, Akimov A V and Dijkhuis J 1 2004 Phys. Rev. Lett. 92 035503.
[8] von Gutfeld R J and Nethercot A H 1964 Phys. Rev. Lett. 12 641.
[9] Stanton N M and Kent A J 2006 Phys. Rev. Lett. 97 115502(R).
[10] Wolfe J P, Imaging Phonons: Acoustic Wave Propagation in Solids (Cambridge University Press, Cambridge 1998).
[11] Saito T, Matsuda O and Wright O B 2002 Physica B316-317 304.
[12] Hao H –Y and Maris H J 2000 Phys. Rev. Lett. 84 5556.
[13] Hao H –Y and Maris H J 2001 Phys. Rev. B64 064302.
[14] Goto T, Yamada-Kaneta H, Saito Y, Nemoto Y, Sato K, Kakimoto K. and Nakamura S 2006 J. Phys. Soc. Jpn. 75 044602.
[15] Kent A J, Stanton N M, Challis L J and Henini M 2002 Appl. Phys. Lett. 81 3497.