Optical excitation and detection of terahertz acoustic waves with semiconductor superlattices

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Abstract. Experimental results about light-to-sound transduction with semiconductor superlattices are presented. Picosecond ultrasonics with pump and probe incident on opposite sides of the substrate allows a full decoupling between generation and detection processes. Associating a metallic transducer, we show that superlattices generate quasi-monochromatic wave packets which can propagate over macroscopic distances in the underlying substrate but are also very selective and sensitive detectors. At the end, using two superlattices simultaneously as phonons generator and detector we evidence that these new transducers allow to perform acoustic experiments at the challenging frequency of 1 THz.

1. Introduction

The realization of very high frequency and monochromatic emitters and detectors is still challenging in acoustics. Terahertz acoustic phonons with wavelength in the nanometer range should be very useful to study vibrational properties of nanostructures, amorphous and quasicrystalline materials at these frequencies. They would be also very interesting for high resolution acoustic microscopy, and for driving acousto-optic devices at the picosecond time scale. Piezoelectric transduction is very efficient up to a few GHz. Reaching the THz range, heat pulses have been used as phonon sources, coupled to superconducting bolometer detectors, but the emitted phonons are incoherent with a broad spectral distribution. Moreover bolometers are sensitive detectors but they are too slow to be able to measure high frequency content of acoustic pulses. Femtosecond laser sources have made possible the generation and detection of coherent acoustic pulses in metallic films with an optical pump and probe technique [1] and picosecond acoustic pulses are generated with a broad frequency spectrum extending up to several hundreds GHz.

Thanks to epitaxial growth it is possible to make semiconductor heterostructures with excellent interface quality and recently many studies report coherent phonon excitation in such structures [2]. We present here experimental results on GaAs/AlAs superlattices performed with the picosecond ultrasonics technique in an original geometry, allowing the decoupling of the generation and detection process. The studied samples and the experimental configuration are presented in section 2. We demonstrate independently that superlattices are quasi monochromatic generator and very sensitive detectors in the 100 GHz–1.2 THz range, by studying samples associating a superlattice on one side and an aluminum film on the other side (section 3). Finally we study samples where a superlattice was
grown on each side of the substrate: one superlattice is used as a phonon generator while the other one acts as a phonon detector. Section 4 presents experimental results and application: in particular we describe an acoustic experiment performed at 1 THz.

2. Samples and experimental configuration
The studied samples are superlattices (SLs) made of a regular stacking of GaAs/AlAs bilayers grown by molecular beam epitaxy on (001) oriented two-side polished GaAs substrates. Due to the periodic modulation of elastic properties between the two materials, acoustic modes are folded in the first Brillouin zone and energy gaps open in the Brillouin-zone center and boundary. Sample A (respectively B and C), having a period of 24.6 nm (respectively 12.5 and 8.3 nm), is designed to have the zone-center first gap located at 200 GHz (respectively 400 and 600 GHz).

The experimental configuration is based on the picosecond ultrasonics technique, which is a pump and probe experiment, in a peculiar configuration where excitation and detection of acoustic waves take place on the opposite sides of a thick substrate [3]. This allows a complete decoupling between the generation and detection mechanisms. The strain pulse is generated by absorption of a femtosecond laser pulse on a transducer (aluminum film or SL) deposited on the backside of the sample. This strain pulse travels through the whole substrate up to the opposite side and is detected by a second transducer (SL or aluminum film) by measuring the reflectivity of the delayed probe pulse. Measurements were performed at 15 K where acoustic attenuation is weak enough to allow propagation through the substrate (330-360 μm). The laser pulses were produced by a femtosecond mode-locked Ti:sapphire laser with a wavelength adjustable from 700 to 800 nm with a 80 MHz repetition rate. The pump was modulated at 1 MHz and the transient changes of the optical reflectivity ΔR/r are measured with a lock-in amplifier. Finally we can access to the real and imaginary part of ΔR/r thanks to a Sagnac interferometer, well suited to the use of the cryostat.

![Figure 1](image_url)

**Figure 1:** Left: Transient reflectivity on sample A in the “generation” configuration, as a function of the time delay. The inset magnifies short time variations. Right: corresponding Fourier transform of the time derivative superimposed to the dispersion curve.

3. Generation and detection

3.1 Generation by a SL
In the “generation” configuration where the pump is focused on the SL whereas the detector is an aluminum film, we study the capability of a SL to generate acoustic waves. On sample A, after the generated acoustic pulse has propagated once through the substrate, we obtain the transient signal plotted in figure 1. It is dominated by a 200GHz oscillation, as can be seen in the corresponding Fourier transform of the time derivative. More precisely, for a weakly absorbing SL, the generation is
expected to be efficient at modes with wave vector $q=0$, and from symmetry considerations, only the lower gap edge mode is excited. The same experiment has been performed at a lower frequency (100 GHz [4]) and also, on sample B, at a higher frequency but only a very weak peak is obtained at 400 GHz in this later case. This result reveals here the lack of sensitivity of aluminum films as detector, as we will see next.

3.2. Detection by a SL
In the “detection” configuration (pump focused on the SL, probe on the metallic film), the pump pulse is absorbed on the metallic film and results in the creation of an acoustic pulse whose spectrum extends up to 0.3 THz. The pump beam has a typical energy of 9 nJ/pulse focused on a 60 μm wide spot, so that non-linear effects occur during propagation into the substrate leading to a pulse distortion and generation of high acoustic frequencies [5]. We take advantage of these non-linear effects to probe the SL detection sensitivity to high frequencies. The Fourier transform of the time derivative of the time traces display a few sharp peaks (figure 2 for sample B). Indeed, as in any photon-phonon interaction, the detection process involves a conservation rule for wave vectors which select phonons with $q=2k$, $k$ being the light wave vector in the SL. This is well verified experimentally as it can be seen by comparing the peak locations in the Fourier transform of the time derivative and the dispersion curve. The peaks are broadened and modulated by Bragg oscillations due to the finite size of the SL. The highest detected frequencies reach 1.2 THz (for samples B and C), which proves how sensitive the SL are. We also performed the same experiment on a sample where the SL is replaced by a 30 nm aluminum film. The detected spectrum, plotted in figure 2, extends only to 350 GHz, and appeared to be much weaker.

![Figure 2](image.png)

Figure 2: Left: Transient reflectivity on sample B in the “detection” configuration as a function of the time delay (laser beams at 780nm). In blue filtered signal at 45GHz, in red at 357GHz. Right: corresponding Fourier transform of the time derivative superimposed on the dispersion curve and a detected spectrum obtained with an aluminum film.

A small oscillation corresponding to Brillouin scattering in the substrate frequency (lowest detected frequency at 45 GHz) starts first and its amplitude suddenly increases (see the arrow) when the acoustic wave front penetrates into the SL, due to the fact that at 780 nm the photoelastic coefficient is larger in the SL than in the GaAs substrate. Later on, the signal still increases and contains beatings of different higher frequencies. It lasts during 500 ps corresponding to the time needed for the acoustic wave to make one round trip in the SL. Filtering the time trace for each detected frequency shows a delay between arrival times of the different frequencies as can be seen from a comparison between filtered signals at the Brillouin frequency and the following detected frequency at 357 GHz. Due to dispersion which occurs during the propagation in the thick substrate, the time delay $\tau$ increases with frequency according to $\tau (\text{ps}) = \tau_0 + 570 \times f(\text{THz})^2$. A systematic analysis of time delays undergone by all detected frequencies was done on sample A. The difficulty is the uncertainty about the distance
from the transducer where such high frequencies are created during propagation and thus about the actual propagation length. However, calculations based on Korteweg-de Vries equation [5] describing nonlinear effects and dispersion during propagation in GaAs show that frequencies up to 600 GHz can be generated after only 30 µm of propagation under our highest excitation which strongly limits the errors.

4. Transduction at higher frequencies

The limitations of aluminium films and the good sensitivity of SL as detector convinced us to study samples associating 2 SLs, one as a generator, the other one to detect the generated acoustic wave, to be able to reach acoustic frequencies in the THz range. We studied a sample containing two nearly identical SL grown at opposite sides of the GaAs substrate. Sample D contains SL with a zone-center first gap located at 1 THz. During the growth process, a thickness gradient was introduced in one of the SL by stopping the wafer rotation; moving the probe beam laser on this SL surface, it was then possible to tune the detection spectral position with respect to the frequency generated by the uniform SL.

Figure 3: Left: Transient reflectivity (a) on sample D as a function of the time delay, filtered signal at the Brillouin frequency (b) and at 1 THz (c). The inset magnifies 1 THz oscillations. Right: corresponding Fourier transform of the time derivative superimposed to dispersion curves of the 2 SLs.

Figure 3 shows the measured signal for the probe optimal position on the wedged SL. The \( q=0 \) mode generated by the uniform SL corresponds to the 2\(^{nd}\) detected frequency satisfying \( q=2k \), the 1\(^{st}\) one being the Brillouin frequency; the 3\(^{rd}\) one is a weaker peak at 1.12 THz. Filtered signals show that 1 THz phonons are detected after an important delay, which we could estimate to be 599±10 ps in good agreement with the expected value (~600 ps).

5. Conclusion

We demonstrated that excitation and detection of Terahertz acoustic waves with femtosecond laser pulses is possible using semiconductor superlattices for transduction. By making use of a configuration where generation and detection processes occur on the opposite sides of a thick substrate and combining metallic films and SL, these two processes have been studied independently. Finally we have shown that acoustic experiments, including generation, propagation over a macroscopic distance and phase detection can be performed at 1 THz.

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