Decoupling of optical generation and detection of acoustic phonons in semiconductor superlattices

M F Pascual Winter, A Fainstein, B Jusserand, B Perrin and A Lemaître

1 Instituto Balseiro & Centro Atómico Bariloche, Av. E. Bustillo 9500, 8400 San Carlos de Bariloche, Río Negro, Argentina.
2 Institut des Nanosciences de Paris, CNRS, Universités Paris 6 et 7, Campus Boucicaut, 140 Rue de Lourmel, 75015 Paris, France
3 Laboratoire de Photonique et de Nanostructures, CNRS, Route de Nozay, 91460 Marcoussis, France

E-mail: pascualm@ib.cnea.gov.ar

Abstract. We present a study of the generation and detection of terahertz acoustic phonons in GaAs/AlAs superlattices through femtosecond pump and probe optical pulses, where the spectral contributions of the generation and detection processes are decoupled and identified. The processes are spatially separated in different superlattices grown with an intermediate 1-µm-thick GaAs layer. Pumping and probing occur on opposite superlattices. A thickness gradient present on one of the superlattices allows us to identify the features in the spectra resulting either from the generation or from the detection. A Raman scattering characterization of the sample is also presented.

1. Introduction

Femtosecond pump-probe experiments have been largely used to study acoustic properties of metals and semiconductors. The generation of acoustic pulses in thin films [1, 2] has been observed via these techniques, and more recently soliton generation and propagation have caught special attention [3]. Ultrafast techniques are excellently suited for analyzing the acoustic vibrations of artificially nanostructured materials, such as superlattices (SLs) [4, 5, 6, 7] -which we will study in this report-, nanocavities [7, 8], quantum dots [9, 10] and quantum wells [11]. The vibrations are generated through the absorption of a pump pulse, and they are optically detected via the changes they introduce in the optical properties of the sample. A probe pulse, whose reflected intensity is measured, reveals the state of the optical properties at a later time. The delay between the pump and the probe pulses is varied in order to obtain the time evolution of the acoustic vibrations.

The spectral analysis of pump-probe experiments performed on superlattices reveal three peaks corresponding to folded acoustic phonons in the region of the first zone-center minigap [4, 12], enclosed by the dashed circle in Fig. 1(a). Among these three peaks, the ones with the lowest and highest energies (empty symbols) can be assigned to modes with \( q = 2k_{pr} \), where \( k_{pr} \) is the probe photon wave vector. The remaining mode (full symbol), not centered with respect to the other two, is identified as the zone-center mode (\( q = 0 \)) of Raman-active symmetry.
detection in the observation of these peaks. Two-color pump-probe experiments have verified that the zone-center mode results from the generation process, while the \( q = 2k_{pr} \) doublet is related to the detection mechanism \([6]\). From the theoretical point of view, the spectral distribution of acoustic modes excited by the pump pulse can be calculated \([13]\). A spectrum with a maximum amplitude at the zone center Raman-active mode is obtained. As regards the detection, the calculation of the spectral response of the change in the probe reflectivity yields maxima at the \( q = 2k_{pr} \) modes \([2, 14]\). Therefore the generation and detection efficiency maxima do not coincide in energy. Nevertheless, due to finite size effects and light absorption some overlap is always assured, and the three peaks are thus observed in the spectral analysis of the pump-probe results.

In the present work we propose a different method for decoupling the generation from the detection. We make use of two SLs separated by a thick intermediate layer along which phonons can propagate from one SL to the other. The vibrations generated by the pump impinging on one SL travel along the intermediate layer until they reach the second SL where the detection takes place. This scheme has already been proposed in Ref. \([13]\). The advantage of our scheme is that one of the SLs presents a thickness gradient, allowing us to identify the contributions from the generation from those of the detections. Furthermore, the bandwidths for the generation and the detection of phonons can be tailored independently, as required, for instance, in the observation of acoustic Bloch oscillations in the terahertz range \([15]\). The energies of the doublet components depend on the SL period \( D \) as well as on the probe wavelength. In the two-color pump-probe experiment the laser wavelength is varied in order to separate both contributions. We propose to explore the possibility of varying the SL period.

2. Results and discussion

The sample is schemed in Fig. 2. It consists of two 60-period GaAs/AlAs SLs separated by a 1-µm-thick GaAs layer. It was grown by molecular beam epitaxy on a [001]-oriented GaAs substrate. For the uniform SL the layer thicknesses are 85 Å/37 Å, according to x-ray diffraction experiments. The other SL was grown with a global thickness gradient around the mentioned values. A central stripe was cut out of the 5-cm-diameter wafer and afterwards cleaved into 17 smaller \( \sim 9 \text{ mm}^2 \) pieces that will be hereafter identified with the numbers 0, ..., 16. Only results for odd number pieces will be presented in this report.

Figure 1. (a) Dispersion relation (acoustic folded branches) for a SL of period \( D \). The full (empty) circles show the generation (detection) maxima.

Figure 2. Scheme of the sample and experimental configuration for the pump-probe experiment.

For characterizing the sample we used Raman scattering, since it is the standard technique for studying folded phonons in SLs and nanostructures in general. We performed room temperature
Raman scattering experiments in a backscattering geometry, with incident radiation from a Ti:sapphire laser. We used a laser power of 5 mW, focused on a ∼50 µm spot. The scattered light was dispersed using a Jobin-Yvon T64000 spectrometer, and detected with a liquid N2-cooled charge coupled device. In order to have enough Raman signal the laser wavelength was tuned to the transition from the second confined electron state to the second confined heavy hole state, in each piece of the sample. The range spanned was thus from 768 nm (piece 01) to 732 nm (piece 15).

Figure 3 shows the Raman spectra for the different pieces (the piece number is indicated to the left of each curve) as well as for the uniform SL. A luminescence background has been subtracted. We observe the lower component of the first doublet (dashed line) and both components of the second doublet (dotted lines). As expected, the phonon modes shift in energy along the wafer. We deduce a thickness gradient of 22%. The uniform SL has a period similar to the one the wedged SL presents in the middle of the wafer. Its lower $q = 2k_{pr}$ mode of the first zone-center minigap is observed at 0.362 THz. It is worth noting that the higher component of that minigap is absent in all the Raman spectra. We relate this fact to resonant character of the spectra. In Raman experiments performed in non-resonant conditions at a wavelength of 514 nm, both components of the doublet were observed.

![Figure 3](image1.png)

**Figure 3.** Raman spectra on both sides of the sample. For the wedged SL the piece number is indicated to the left of each curve. The dashed line follows the lower mode of the first zone-center minigap doublet. The dotted lines follow the second zone-center doublet.

![Figure 4](image2.png)

**Figure 4.** Fourier transforms of the time dependence of $\Delta R/R$. The spectra are labeled with the sample piece number. The starred peak is an artifact due to the laser power stability system. The arrows indicate the expected position of the generation peak and the vertical line shows the position of the detection one.

We performed room temperature reflection-type pump-probe experiments at a central optical pulse wavelength of 800 nm, both for the pump and the probe. Femtosecond pulses (≈80 fs) from a mode-locked Ti:sapphire laser, with a repetition rate of 79 MHz, were split into the pump (energy 1.5 nJ) and probe (0.22 nJ) pulses. The pump was modulated at 1.8 MHz by an acousto-optic device in order to allow for synchronous detection. Both pulses were focused onto ∼50 µm-diameter spots, the pump on the wedged SL and the probe on the uniform SL.

The Fourier transforms of the time dependent change in the probe reflectivity ($\Delta R/R$) are
plotted in Fig. 4. Each spectrum is labeled with the sample piece number as in Fig. 3. In the original data, that is $\Delta R/R$ as a function of time, at short times the signal displays oscillations due to phonons generated in the probe SL by the portion of the pump pulse that is not absorbed in the pump SL, or alternatively, due to phonons detected in the pump SL because of some penetration of the probe pulse. To reduce these contributions, the Fourier transforms have been integrated over times longer than 210 ps, that is the travel time for the phonons to go through the intermediate GaAs layer. We observe a peak fixed along the wafer at an energy of 0.360 THz (vertical line). By comparison to the Raman spectra this peak can be identified with the detection maximum, i.e. the $q = 2k_{pr}$ mode of the uniform SL. The zone-center modes are not accessible via Raman experiments in a backscattering geometry. Nevertheless, their energies can be deduced from those of the $q = 2k_{pr}$ mode and the elastic properties and refractive indexes of GaAs and AlAs. We have marked the expected positions for the Raman-active $q = 0$ mode with the arrows in Fig. 4. We see that the arrows fall very close to a peak that shifts along the wafer. This peak is thus the generation maximum. The difference between the expected energies and the actual position of the peaks might be due to the fact that the estimation made from the $q = 2k_{pr}$ modes is valid for an infinite SL. For sample pieces 01 and 03, the generation and detection peaks are close to each other and some enhancement of the signal is observed.

3. Conclusion
In conclusion, we have decoupled the generation and detection of acoustic phonons in a pump-probe experiment. For that, we have spatially separated both mechanisms in two different SLs grown with an intermediate GaAs layer. Thanks to a thickness gradient present in one of the SLs, we have been able to identify the contributions of the generation and detection processes in the spectral analysis of the pump-probe results, with the help of a Raman scattering characterization.

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