Coherent acoustic phonons in phonon cavities investigated by asynchronous optical sampling

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Abstract. Using a recently introduced measurement technique, called asynchronous optical sampling (ASOPS), we have investigated the dynamics of coherent acoustic phonons in a semiconductor heterostructure composed of a GaAs film between two GaAs/AlAs superlattices, serving as a cavity for acoustic phonons. Measurements were performed at liquid helium temperatures. The possibility to perform two-color pump-probe spectroscopy allowed us to tune the probe pulse energy to the cavity band gap, while sweeping the pump pulse energy over the superlattice resonance. The large measurement window of 1 ns in combination with a resolution of about 150 fs made a detailed analysis of the observed phonon dynamics possible. We observed a long lived oscillation in the gap of the phonon dispersion at 466 GHz, which we attribute to a cavity mode.

1. Introduction
Coherent phonon oscillations in a variety of materials including bulk semiconductors and semiconductor heterostructures have been studied for some time now [1,2,3]. Technological advancements, concerning mainly the femtosecond laser systems, have contributed to a rising relevance of this field for both fundamental research and technological application. In recent years a wide variety of applications have been demonstrated e.g. in the semiconductor industry for metrology. Different methods have been presented to investigate the characteristics of phonon pulse generation and detection, including the possibility to coherently control the phonon pulses [4,5,6,7]. The interaction of photons and phonons plays a major role in all of these applications, since the generation and detection processes rely on ultra-short laser pulses. Recent results demonstrate the possibility to enhance this interaction, by confining the phonon oscillations and the light pulses locally in phonon cavities. In such a structure a single acoustic cavity mode can couple to an external electromagnetic field very efficiently, generating as a result coherent monochromatic phonons [8,9,10]. Such a device could be used for example as a basis for a phonon laser [11].

These cavities are built using superlattices in such a way, that they act as acoustic mirrors much like in optical micro cavities. If the superlattice is made of two materials with different impedances, the additional periodicity leads to a reduced Brillouin zone (BZ) with a maximal wave vector \( q_{\text{max}} = \pi/d_{\text{SL}} \), where \( d_{\text{SL}} \) is the superlattice period. The phonon dispersions are folded back into the reduced BZ giving rise to phonon frequencies of a higher order near the zone center. If the relation of the thicknesses of both materials is chosen correctly, acoustic minigaps open up at the center and at the
edge of the BZ. A cavity mode with a frequency within the minigap can be trapped between two superlattices acting as mirrors.

2. Asynchronous optical sampling (ASOPS)

In this paper we have analyzed such a cavity sample via asynchronous optical sampling (ASOPS). ASOPS is a variation of classical pump probe spectroscopy, where two femtosecond lasers with a repetition rate of about 1 GHz are implemented (Gigajet TWIN, Gigaoptics GmbH, Germany), one serving as the pump source, the other one as the probe source. Both lasers are tuned in such a way, that the repetition frequency of the pump laser is exactly 10 kHz higher than the repetition frequency of the probe laser. This difference is stabilized via an active feedback phase locked loop. Consequently the time delay between pump and probe pulse is automatically swept between zero and the maximum time delay given by the inverse of the repetition frequency, while the time needed to perform this sweep is given by the inverse of the difference repetition frequency. Our present configuration allows for a 1 ns time window swept in 100 μs with a time resolution throughout the entire window below 180 fs, leading to a very high signal to noise ratio of 60 db in 60 seconds acquisition time. Both lasers are independently tunable between 1.46 eV and 1.65 eV with a spectral pulse width of approximately 20 nm, allowing for two color pump probe spectroscopy. Further details concerning the measurement system can be found elsewhere [12].

The sample consists of a GaAs film with a thickness of 100 nm serving as a wide cavity enclosed by two GaAs/AlAs superlattices each one consisting of 20 periods of 7.9 nm (28 monolayers) of GaAs and 2.8 nm (10 monolayers) of AlAs. The entire structure is grown on an AlGaAs buffer layer on a GaAs substrate using molecular beam epitaxy. At a temperature of 10 K the first interband resonance of the superlattice is located at about 1.58 eV while the resonance of the GaAs cavity is 70 meV below at 1.51 eV. Experiments shown in this paper were performed at 10 K using a constant probe pulse energy of 1.53 eV, i.e. close to the cavity resonance. The energy of the pump pulse is swept over the resonances mentioned above.

![Figure 1](image-url) Exemplary transient showing the folded phonons mode extracted from the ΔR/R signal shown in the inset by a moving average. This transient was obtained with a pump energy of 1.64 eV.
3. Experimental results

Figure 1 shows an exemplary transient that was obtained at 10 K with a pump pulse energy of 1.64 eV. The observed oscillations are extracted from the original data via a moving average, i.e. averaging over adjacent data points with a temporal window of the oscillation period, to eliminate contributions from the electronic response and the Brillouin branch oscillations at about 40 GHz. One can clearly see a beating as a result of the different spectral components in the signal.

The spectral information is displayed in Fig. 2 a) and b). The lower part of Fig. 2 a) shows the Fourier transform of the transient from Fig. 1 in a range between 250 and 550 GHz, while the upper part shows the corresponding part of the folded phonon dispersion, calculated using the Rytov model, clearly revealing the minigap at the zone center between 449 and 473 GHz. The solid grey arrows show the frequencies, where peaks in the phonon spectrum are expected, corresponding to the lower $q_{\text{phonon}} = 0$ branch and $q_{\text{phonon}} = 2k_{\text{probe light}}$ modes. No peak is expected at the upper $q=0$ branch since it is Raman forbidden because of its symmetry. It is clearly visible that there is a very good agreement with the peak positions of the experimental results. The fine structure of the spectral data can be very well explained with finite size effects due to the limited amount of periods in the superlattice and interference effects due to the double superlattice structure [13]. The dashed gray lines and corresponding arrows show the positions for the resulting discrete frequencies in the phonon dispersion. The splitting of both 2k-modes again agrees well with the theoretical prediction.

![Figure 2](https://example.com/figure2.png)

**Figure 2.** (color online) Spectral information of the phonon modes of interest. a) compares the spectrum obtained at 1.64 eV with the calculated phonon dispersion in the upper part of the figure while b) shows a series of spectra obtained at different pump pulse energies. The solid grey arrows mark the position of the $q=0$ mode and the two 2k-modes given by the intersection of the dispersion curve and the dashed-dotted line corresponding to twice the wave vector of the probe light. The dashed grey lines in the upper part of a), indicate the positions of the discrete frequencies in the phonon dispersion due to finite size effects. The short black arrows in both parts a) and b) indicate the cavity mode.

Fig.2 b) on the other hand shows a series of spectra taken at different pump pulse energies from 1.64 eV well above the resonances of the superlattice and the GaAs cavity to 1.47 eV, i.e. below the GaAs cavity resonance. The peak intensity decreases for all three modes ($q=0$ and the two $q=2k_{\text{probe light}}$) with decreasing pump pulse energy. The $q=0$-mode, which almost disappears completely...
at 1.47 eV, decreases much faster than both 2k-modes. But even when the pump pulse energy lies underneath the superlattice resonance energy, a clear phonon spectrum is visible. This is a clear indication for non-resonant Raman scattering as the generation process.

In addition to the q=0-mode and both 2k-modes there is another peak in the phonon spectrum located at 466 GHz, which lies exactly in the minigap of the phonon dispersion marked by the black arrow in Fig.2. The wavelength of this mode fits exactly 10 times into the 100 nm thick cavity. In spectra of single superlattices no such modes in the gap were observed. The intensity of this gap mode also decreases with increasing wavelength, disappearing almost completely at a pump pulse energy of 1.47 eV. We attribute this peak to a cavity mode in the GaAs film between the two superlattices. Because of the large thickness of 100 nm in comparison with the SL layers, the electron and phonon dynamics can be described by the dynamics in bulk GaAs. This means, that the phonon generation occurs through the relaxation of photoexcited electrons into LO phonons, which then will decay into acoustic phonons through anharmonic decay with a time constant of 10 ps [14]. This process will generate phonons of all wave vectors, from which only a very small fraction is trapped inside the cavity. As a consequence of this model, this peak should start to disappear, once the pump pulse energy lies below the bulk gap energy plus the LO phonon energy, which occurs at about 1.56 eV. The experimental results displayed in Fig.2 b) indeed show, that the cavity peak strongly decreases at about this energy.

4. Conclusion
The phonon dynamics of a superlattice cavity structure were studied using asynchronous optical sampling. The theoretical predictions for the spectral positions of the q=0-mode and both 2k-modes matched the experimental results very well. The spectra are strongly affected by finite size effects and interference effects due to the double superlattice structure. In addition to the superlattice phonon modes, a further mode, lying in the minigap of the superlattices has been found. We attribute this peak to a cavity mode generated by the relaxation of LO phonons into LA phonons in the wide GaAs cavity.

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