High-speed asynchronous optical sampling for high-sensitivity detection of coherent phonons

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Abstract. A new optical pump-probe technique is implemented for the investigation of coherent acoustic phonon dynamics in the GHz to THz frequency range which is based on two asynchronously linked femtosecond lasers. Asynchronous optical sampling (ASOPS) provides the performance of an all-optical oscilloscope and allows us to record optically induced lattice dynamics over nanosecond times with femtosecond resolution at scan rates of 10 kHz without any moving part in the set-up. Within 1 minute of data acquisition time signal-to-noise ratios better than $10^7$ are achieved. We present examples of the high-sensitivity detection of coherent phonons in superlattices and of the coherent acoustic vibration of metallic nanoparticles.

1. Introduction

The characterization of multilayered thin films via laser-based ultrasound has been a well-established technique since the late 1980s [1,2]. In laser-based ultrasound experiments ultrashort laser pulses generate a strain pulse in an absorbing layer. The propagation of this strain pulse is monitored through changes of the reflectivity of the sample with a time delayed probe pulse. In multilayers such as semiconductor superlattices (SLs) acoustic lattice modes, i.e. zone-folded acoustic phonons in the THz frequency range, can be generated coherently and their propagation and decay dynamics can be studied in the time domain [3-5]. An active field of research is the preparation of cavities for acoustic phonons in SLs in order to study the dynamics of confined acoustic phonon modes [6,7]. Other systems, where the dynamics of coherent acoustic excitation provide new insight into the energy dissipation after pulsed optical excitation, are metallic nanoparticles [8-10]. All these investigations have in common that reflectivity changes or transmission changes need to be monitored with a sufficiently high signal-to-noise ratio over typically several 100 ps of time delay. In conventional pump-probe experiments, the time delay between the pump and probe pulses is accomplished via a mechanical delay line. This approach has several inherent drawbacks: i) a mechanical delay line is limited in speed, ii) it is always a source of mechanical vibration, and iii) it may introduce pointing variations of the laser spot and changes in the spot size. These disadvantages can be circumvented through asynchronous optical sampling (ASOPS) [11-13]. In ASOPS two different femtosecond lasers are employed which provide the trains of pump and probe pulses, respectively. The repetition rate of these lasers is stabilized at an off-set frequency in the kHz frequency range. Hence the maximum time delay given by the inverse repetition rate of one of the lasers is scanned with the off-set frequency.
The achievable time-resolution is defined by the detailed values of the frequencies involved and the detection hardware as explained later. Such a system does not include a moving part in the set-up except a piezoelectric transducer in one of the laser cavities for the stabilization of the repetition rate.

2. High-speed asynchronous optical sampling

While first implementations of ASOPS were based on pulsed lasers with repetition rates below 100 MHz [11], we present a high-speed ASOPS scheme based on two femtosecond lasers with 1 GHz repetition rates (Gigajet TWIN, Gigaoptics GmbH, Germany) [13]. Both lasers are Kerr-lens modelocked Ti:sapphire lasers which are based on a ring cavity with chirped mirrors for dispersion control. One laser serves as master laser and its repetition rate is measured with a fast photoreceiver. The other laser is operated at a repetition rate off-set by 10 kHz with respect to the master oscillator. Both lasers provide pulses around 30 fs duration, up to 1 W average power and independent wavelength tunability between 730 nm to 850 nm.

Figure 1 depicts the schematic sketch of the high-speed ASOPS system. A portion of the probe oscillators pulse train is detected with a fast Si-pin photodiode (PD1, 3 GHz bandwidth, S7911, Hamamatsu Photonics, Japan) to obtain an electronic signal at the repetition rate $f_1$. The signal contains also higher harmonics of the fundamental repetition frequency. The signal is split into a branch at the fundamental repetition rate and a branch at the third harmonic using a power splitter and bandpass filters. The third harmonic signal is upshifted by 30 kHz with a frequency shifter. The frequency shifter consists of a single sideband (SSB) generator and an additional circuit that cancels leakage of the input signal through the SSB generator. This is accomplished with a small portion of the SSB generator input which is split off using a directional coupler and is added to the output with of a microwave combiner with a 180° phase shift. Contributions at the carrier are suppressed by better than 50 dB. The pump oscillator’s pulse train with repetition rate $f_2$ is detected with a second photodiode (PD2) whose signal is also split into the fundamental and the third harmonic. The third harmonic of $f_2$ is phase-locked to the upshifted third harmonic of $f_1$ with a double-balanced mixer (DBM) serving as phase detector. The DBM output is supplied to a piezo-electric transducer (PZT) that supports a cavity mirror of the pump oscillator via a proportional-integral amplifier and a high voltage amplifier. The effective bandwidth of the entire feedback loop is 5 kHz determined by the feedback bandwidth of the PZT-mirror assembly (TL-1000, Gigaoptics GmbH, Germany). The off-set frequency between $f_1$ and $f_2$ is fixed to 10 kHz. The maximum time delay between two successive pulses of approx. 1 ns is scanned within 100 $\mu$s in real-time. The signals at $f_1$ and $f_2$ from the fundamental signal branches are fed into a second DBM whose output is converted to a 10 kHz TTL-level signal which serves to trigger the data.

Figure 1. Schematic sketch of ASOPS set-up. The solid black lines represent the optical beam path, the dashed black line electronic signals. PD is a photo diode and DBM is a double balanced mixer.
acquisition. The time resolution of the ASOPS set up is given by the ratio of the off-set frequency (10 kHz) and the product of the repetition rate (1 GHz) and the detection bandwidth (100 MHz), i.e. 100 fs in our case. The same system with 100 MHz repetition rate lasers would have a time resolution of 1 ps at the same off-set frequency. Hence lower off-set frequencies would be required for achieving a 100 fs time-resolution. A further reduction of the time resolution is introduced by timing jitter, which is dependent on the accumulated time delay between pump and probe pulses [13]. The overall time-resolution is better than 160 fs, which is sufficient for most investigations of coherent acoustic phonon dynamics.

3. Coherent acoustic phonon dynamics in superlattices

The investigation of coherently excited acoustic phonons in semiconductor SLs and quantum wells has been a subject of intense research throughout the last decade in different material systems such as GaAs/AlAs [3,5] and InGaN/GaN [4]. Pump-probe experiments provide a direct access to the investigation of propagation and dephasing dynamics. In the following we like to demonstrate the sensitivity obtained with high-speed ASOPS on a standard GaAs/AlAs SL under excitation conditions in resonance with the first heavy-hole to electron interband transition and on InAs/GaSb SLs excited off-resonance far above the fundamental interband transition.

Figure 2 depicts the time domain traces, the extracted oscillatory contribution and the Fourier transform of a 40 periods (GaAs)$_{19}$(AlAs)$_{19}$ SL (the index is the number of monolayers) obtained within 100 s of data acquisition. The first and second order zone-folded modes at q=0 and q=2$k_{probe laser}$ are clearly resolved. The linewidth of the Fourier peaks is 3.8 GHz, which is still larger than the frequency resolution of 1 GHz of our set-up. The corresponding decay time of the q=0 mode at 450 GHz is 300 ps. This lifetime is in agreement with the one for q=0 modes observed by Trigo et al. [5]. The smaller peaks in the spectra stems from the finite size of the SL with 40 periods.

![Figure 2](image-url)

**Figure 2.** a) Time domain trace of reflectivity changes of a (GaAs)$_{19}$(AlAs)$_{19}$ SL under resonant excitation and detection conditions. b) extracted oscillatory structure of the signal in a). The inset shows a zoom into the data between 60 ps and 110 ps. c) Numerical Fourier transform of time domain data in b).
The investigation of zone folded acoustic phonons in InAs/GaSb SLs is of interest since the two compounds do not possess neither a common anion or cation. Hence, depending on the growth sequence the interface can be GaAs-like or InSb-like. Samples with InSb-like interfaces exhibit narrower X-ray SL diffraction peaks than those with GaAs-like interfaces indicating a higher layer quality [14]. Figure 3 depicts the oscillatory part of time-resolved reflectivity changes of (InAs)$_{14}$(GaSb)$_{10}$ SLs with different interfaces obtained for excitation and probe energies of 1.6 eV, i.e. far above the fundamental transition of the InAs wells. The Fourier transforms of the time-domain data show a modulation at the Brillouin branch frequency and a triplet at the first order close to $q=0$ zone-folded modes. No higher order modes are resolved as in the Raman scattering spectra of the same samples [14]. However, in the Raman spectra in backscattering geometry only a doublet according with $q=2k_{\text{max}}$ has been observed. The decay times (linewidths) of the coherent phonons are significantly shorter (broader) than those in the GaAs/AlAs SL. Also the decay time of the sample with InSb-like interfaces is longer than for the one with GaAs interfaces, indicating that the interface quality and/or strain of the samples is reflected in the acoustic phonon dynamics. The peak position of the phonon frequencies matches very well the theoretical calculation of the dispersion curves for the SLs with InSb-like interface, while the sample with GaAs-like interface are shifted to higher frequencies [14]. This indicates an influence of the interface on the sound velocities, since GaAs has a higher sound velocity than InSb. Interestingly, we also observed a mode at the zone-edge below 300 GHz. Such zone-edge modes have been observed only for large-period SLs, i.e. when the light wave vector is larger than the maximum wave vector of the mini-Brillouin zone. This leads to the observation of umklapp-mediated Raman lines [15,16]. Since this is not the case in our samples the observation requires either a detection process sensitive towards a zone-edge excitation or a modulation of the SL optical properties at a period of 2 $d_{\text{SL}}$.

Figure 3. a) Time domain trace of reflectivity changes of a InAs/GaSb with InSb-like interface (left) and GaAs-like interface (right). Please note the change in the time scales depicted. b) numerical Fourier transforms of the data in a).
4. Coherent vibrational dynamics of gold nanoparticles

Nanoparticles show confined phonon modes in similar way as artificial multilayered crystals as above. For nanoparticles these modes have no dispersion, while a number of harmonics related to the symmetry conditions in angular or radial direction are observed [17]. In the past the understanding of these modes, in particular the optically detected fundamental Lamb mode helped to gain insight into the interactions of the electron and phonon subsystems or elastic properties of the solid phase in particular under nanoconfinement [18, 19].

ASOPS was applied to the investigation of gold particles that are organized as dilute monolayer on a silicon substrate. By pumping and probing in the near infrared, the fundamental vibrational mode could be resolved to high precision. At the same time, the speed of the measurement process allowed to vary external parameters in-situ and track the ultrafast response. While scanning the temperature of the system a discontinuity in the elastic properties was observed, that points towards a surface phase transition of the particles [10]. Within a narrow temperature interval these oscillations abruptly change the amplitude, the damping rate and also the phase relative to the excitation. This observation is consistent with a phase transition involving damping through a viscous surface layer of few nanometers thickness.

![Figure 4](image_url)

**Figure 4.** Grey scale plot of the transient periodic reflectivity change of a 60 nm gold particle layer during the variation of the external temperature. The general transient response includes a sharp sub-picosecond peak related to the electron gas heating and cooling and a baseline extending to hundreds of picoseconds representing the thermal response and cooling process (inset, logarithmic scale). Superposed are the undulations of the reflectivity at 795 nm, which are caused by the periodic volume change of the particles connected to the fundamental Lamb mode.

5. Conclusions

High-speed asynchronous optical sampling (ASOPS) provides new capabilities for the time-resolved detection of GHz to THz phonon dynamics in pump-probe spectroscopy. This technique enables us to monitor reflectivity changes in the order of $10^{-7}$ in less than a minute of data acquisition time with 100 fs time-resolution over 1 ns of time-delay. Hence this technique enables to perform material characterization and investigation of fundamental questions related to coherent acoustic phonons with significantly improved performance as compared to pump-probe spectroscopy employing mechanical delay lines. Two-color experiments provide a large flexibility for the investigation of a variety of systems through separate tuning to electronic resonances.
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