Time-and-Wavelength Resolved Pump Probe Spectroscopy & Calibration Measurement Using Semiconductors

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Pump-probe experimental setup using 10 fs ultra-short laser pulse was successfully developed. Using this setup, the excitation of coherent phonons in Te and GaAs were measured for calibration and accuracy test of the developed setup. Coherent Raman-active phonons are observed by PP experiments. 10 fs pulse was not necessary to generate coherent optical phonon. Strongly chirped pulse with roughly ~ 20 fs duration can generate it. Frequencies of these modes was in good agreement to values reported. It was demonstrated how spectrum of laser near of band-gap could significantly affect the pump-probe signal. In near of band-gap, the PP response of GaAs is dependent on the spectrum of the laser, but coherent optic phonon oscillation isn't dependent on it. Time-and-frequency resolved pump-probe optical spectroscopy is used to investigate the effect of spectral dependence of pump-probe of GaAs. The measured modifications of the optical properties of GaAs are consistent with the creation of a non-thermal state, metastable on the ps timescale, after the pump-induced impulsive modification of the electron interactions.

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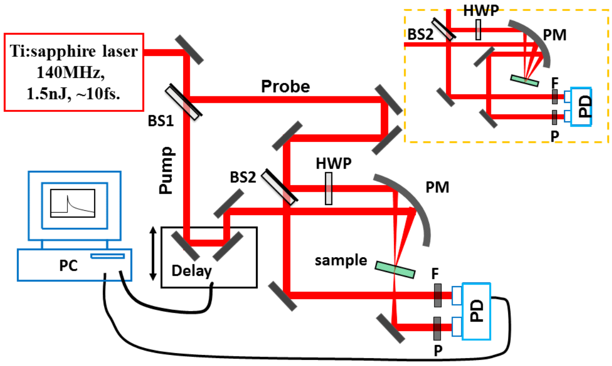
I. INTRODUCTION

Since the early 1980's, many groups have been studying the response of material systems to optical pulses on a femtosecond time scale using optical pulse pump-probe (PP) techniques. In a number of these experiments, oscillations due to a Raman excitation process are observed in the transmission of probe pulses as a function of time following the exciting pump pulse. Recently, femtosecond PP techniques have been applied to the study of metals and superconductors. In PP experiments on a number of conducting or semiconducting materials, oscillations have been observed in reflectivity (or transmission through thin samples) with frequencies that correspond to optical phonon modes of the samples. A number of mechanisms have been proposed to explain these oscillations, including impulsive stimulated Raman scattering , a nonlinear optical susceptibility mechanism, and the screening of space-charge fields at the surface of semiconducting samples.

In this paper, we developed pump-probe experiment which is appropriate for investigation of non-equilibrium system. In II section we will introduce the experimental setup and the laser source which is used to PP experiment.

In III section, a PP reflectivity of a GaAs single crytal is measured to check the measurement system. The PP data for GaAs was already reported in literature. G.C.Cho *et al.* measured PP reflectivity changes for GaAs as a function of delay time between pump and probe pulse using 50 fs laser pulses from a ring dye laser [2]. The PP response is strongly dependent on the pulse wavelength near the band gap of GaAs.



Зураг 1: Pump-probe transmittance measurement setup. PM: parabolic mirror, BS1: 70:30 beam splitter, BS2: 50:50 beam splitter, P: polarizer, F: neutral density filter. PD: photo detector, HWP: half wave plate. The inset is pump-probe reflectance measurement setup.

In IV section, we reported coherent phonon oscillations in Te crystal which is known as a good test system. It is because the coherent oscillation appears very clearly even with weak laser intensity.

In V section, we discussed the main difference of the mode-locking condition dependence of PP reflection signal reported in III section.

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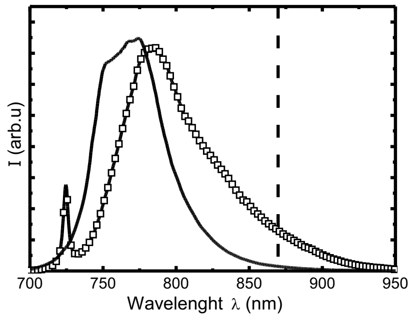


Зураг 2: Spectrums of femtosecond laser. Vertical dashed line marks the band gap of GaAs.

II. EXPERIMENTAL SETUP

For the PP measurement, the laser pulses were generated from a mode-locked Ti:sapphire oscillator [1]. The center wavelength of the pulse was 800 nm (1.5 eV). The pulse duration was 10 fs. The pulse energy was ~ 1.5 nJ at a repetition rate of 140 MHz. Experimental setup was shown in Fig. 1.

The band width of the laser pulse was shown in Fig. 2. The laser beam was split into a pump beam ($\sim 75\%$) and a probe beam ($\sim 10\%$). The pump and probe beams were focused on the surface of a sample with $100\,\mu m$ in diameter. The pump beam was normally incident to the surface, the angle between the probe beam and surface normal was approximately 8°. The transmitted or reflected probe beam was detected with a balanced photo-detector, which subtracted the intensity of a reference beam to eliminate the laser noise. The temporal coincidence of the probe and pump pulses was checked by the sum frequency signal generated from a β-$BaBO_2$ crystal. The zero-time-delay was defined as the time delay point where the sum frequency signal was strongest. All experiments were performed at room temperature.

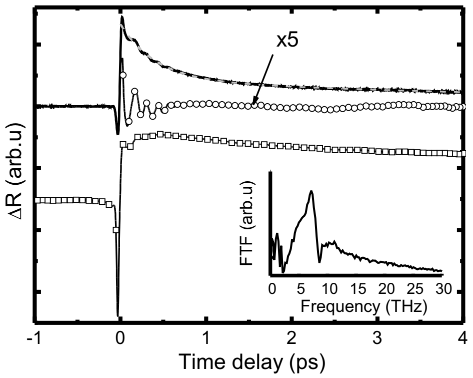
The inset of Fig. 1 shows the PP reflectance measurement setup. Here the reflected probe pulse from the sample surface were collimated by the parabolic mirror and guided by flat mirrors to the balanced detector. Others are same with the transmittance measurement setup.

III. CALIBRATION OF RELAXATION BEHAVIOR AND PHONON OSCILLATIONS USING *GaAs* CRYSTAL

A PP reflectivity of a GaAs single crytal was measured to check the measurement system. The PP data for GaAs was already reported in literature. G.C.Cho *et al.* measured PP reflectivity changes for GaAs as a function of delay time between pump and probe pulse using 50 fs laser pulses from a ring dye laser [2]. The polarization of the probe beam was kept orthogonal to the pump beam. Periodic LO-phonon oscillation was only visible, when the polarization of the pump beam was parallel to the [011] or [01$\bar{1}$] axis, as required by the selection rules for RS . The frequency of this periodic oscillation matched exactly the frequency of the LO phonon at $\vec{q} = 0$ in GaAs (8.8 THz).

In the thesis experiment, the PP response is strongly dependent on the pulse wavelength near the band gap of GaAs (1.424 eV or 870 nm). Spectrums used in the experiments were shown in Fig. 2 for two deferent pulses. Vertical dashed line shows band gap of GaAs. One spectrum was whole in region of above band-gap (solid line). Some part of another spectrum was in under band gap (open square line).

In Fig. 3, the solid and open square lines shows the PP reflectivity changes in GaAs for two deferent pulses, respectively. The response of the system shows small amplitude coherent oscillations, superimposed on an exponential decay. For spectrum with above band-gap, the decay was fitted well with a double exponential of the form $\sum_{i=1,2} A_i e^{-t/\tau_i}$ (gray dashed line). The first term is attributed to rapid electron-electron thermalization, $\tau_1 = 150$ fs. The slower, $\tau_2 = 2.5$ ps, second term is attributed to electron-phonon thermalization. In Fig. 3, the open circle line shows the coherent oscillations after the electronic backgrounds have been subtracted. The Fourier transforms of the data (open circle line) is shown in the inset of Fig. 3. Periodic LO-phonon oscilla-



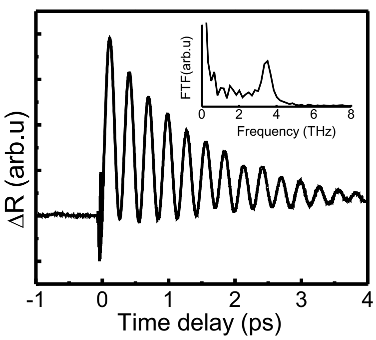
Зураг 3: Pump-probe reflectance change of GaAs.

tions are only visible, when the polarization of the pump beam is parallel to the [011] or [01$\bar{1}$] axis. The dominant oscillation have frequency of 8.8 THz and correspond, with good agreement to the values reported in measurements reported by Cho *et al.* [2] and Sadao Adachi [3]. The consistency demonstrated that the setup of Fig. 1 worked properly. To check pulse spectrum dependence, we developed frequency resolved PP experiment in section V.

IV. CALIBRATION OF PHONON OSCILLATIONS USING *Te* CRYSTAL

Te crystal is known as a good test system to observe the coherent phonon oscillations using laser pulse pump-probe experiment. It is because the coherent oscillation appears very clearly even with weak laser intensity. Coherent lattice vibrations in Te were first reported by a group at MIT [4]. Using a single-color PP setup, oscillations in the reflectivity were observed at the frequency of the symmetry-preserving A_1 phonon mode (3.6 THz) and at no other after excitation by a 60 fs pulse of influence $2.5\,\mathrm{J/m^2}$. Although the excitation of the symmetry-preserving A_1 phonon mode in Te dominates the optical response, the coherent excitation of other phonon modes has also been observed [5, 6]. The observation of oscillations at 2.77 THz and 4.2 THz in the anisotropic reflectivity, precisely the frequency of the two E_{TO} phonon modes in Te, suggested that other modes can be excited. Anisotropic reflectivity changes in other geometries as well as the detection of THz emission from Te under short-pulse excitation revealed that coherent E_{LO} phonons can be excited.

Fig. 4 depicts the PP reflectivity change from Te surface as a function of time delay (denoted as isotropic reflectivity change). The Fourier transforms of the time domain data are shown in the inset. The isotropic reflectivity change is strongly modulated by the coherent excitation of the fully symmetric A_1 mode at 3.6 THz, which is selectively excited via the displacive mechanism [2, 7]. This value is also good agreement to value reported by Dekorsy *et al.* [5], which demonstrated that the set-up of Fig. 1 worked properly for coherent phonon measurement.



Зураг 4: Pump-probe reflectance change of Te. In the inset, the Fourier transform of the data, showing oscillation at 3.6 THz.

In conclusion, a pump-probe experimental setup using 10 fs ultra-short laser pulse was successfully developed. Using this setup, the excitation of coherent phonons in Te and phonons and relaxation behavior in GaAs were measured for calibration and accuracy test of the developed setup. Experimental results consistent with the data in literature were obtained, which demonstrated that the setup worked properly and accurately.

V. WAVELENGTH OR FREQUENCY RESOLVED PUMP-PROBE EXPERIMENT IN *GaAs*

In this section, we explain the main difference of the mode-locking condition dependence of PP reflection signal discussed in section III. In order to obtain photon energy resolution, probe pulse was dispersed through a diffraction grating and detected after reflected on sample. The response was strongly dependent on the probe wavelength near the band gap of GaAs. The diffraction grating was located along the optical line of the reflected signal: it is in charge of the dispersion of the reflected spectrum before it is acquired by a photodiode. A slit in front of the photodiode was used for selecting a thin range of wavelengths of the spectra. The diffraction grating was setup on a rotation stage. Selected wavelength range can be changed by rotating of the diffraction grating. A spectrometer was used to check wavelength selection before the measurement.

We measured the normalized variation of reflectivity

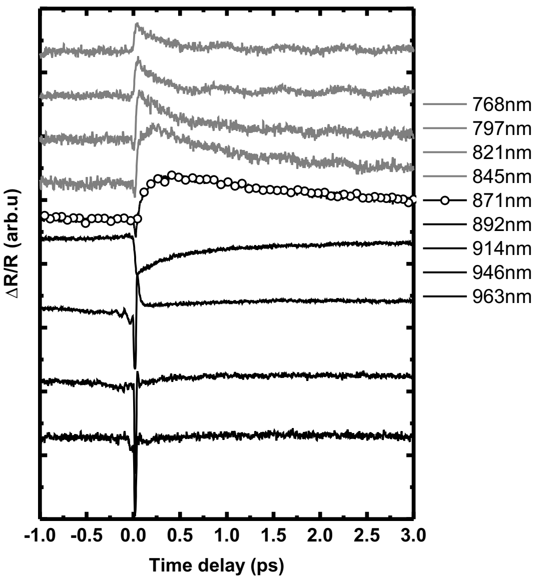
$$\Delta R(\lambda,\tau) = \frac{R(\lambda,\tau) - R(\lambda,-\infty)}{R(\lambda,-\infty)}, \tag{1}$$

where τ is the delay between the two laser pulses, $R(\lambda,\tau)$ is the reflectivity of the pump excited sample, $R(\lambda,-\infty)$ is the reflectivity in the equilibrium state or before pumping. λ is wavelength of the selected thin range.

The obtained raw data was shown in Fig. 5. Laser pulse was broad spectrum in Fig. 2 (open square line).

As Fig. 5, $\Delta R(\lambda,t)$ above the band gap was similar to the solid lines in Fig. 3. However, in $\Delta R(\lambda,t)$ below the band gap, the amplitude (A) of rapid electron-electron thermalization was negative sign. Near of zero time delay, frequency resolved PP signal was strongly dependent on wavelength. The open square symbol line in Fig. 3 was the sum contribution of these two kind of signals.

In conclusion, a pump-probe experimental setup using 10 fs ultra-short laser pulse was successfully developed. Using this setup, the excitation of coherent phonons in Te and GaAs were measured for calibration and accuracy test of the developed setup. Coherent Raman-active phonons are observed by PP



Зураг 5: Wavelength resolved pump-probe signal $\Delta R(\lambda,t)/R$ of GaAs crystal.

experiments. 10 fs pulse was not necessary to generate coherent optical phonon. Strongly chirped pulse with roughly ~ 20 fs duration can generate it. Frequencies of these modes was in good agreement to values reported. It was demonstrated how spectrum of laser near of band-gap could significantly affect the pump-probe signal. In near of band-gap, the PP response of GaAs is dependent on the spectrum of the laser, but coherent optic phonon oscillation isn't dependent on it.

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