DEVELOPMENT OF PHONON DYNAMICS MEASUREMENT SYSTEM BY MIR-FEL AND PICO-SECOND LASER

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Abstract

Coherent control of a lattice vibration in bulk solid (mode-selective phonon excitation: MSPE) is one of the attractive methods in the solid state physics because it becomes a powerful tool for the study of ultrafast lattice dynamics (e.g. electron-phonon interaction and phonon-phonon interaction). Not only for that, MSPE can control electronic, magnetic, and structural phases of materials. In 2013, we have directly demonstrated MSPE of a bulk material with MIR-FEL (KU-FEL) by anti-Stokes Raman scattering spectroscopy. For the next step, we are starting a phonon dynamics measurement to investigate the difference of physical property between thermally excited phonon (phonon of equilibrium state) and optically excited phonon (phonon of non-equilibrium state) by time-resolved method in combination with a pico-second laser and MIR-FEL. By using pico-second laser, we can also expect to perform the anti-Stokes hyper-Raman scattering spectroscopy to extend MSPE method to the phonon mode which has Raman inactive (or some of the infrared inactive modes). As the first step, we have commissioned the time-resolved phonon measurement system and started the measurement on 6H-SiC. Consequently, we succeeded in a development of a phonon dynamics measurement system and the temporal resolution of the developed system was around 10 ps.

INTRODUCTION

The electron-phonon interaction influences physical properties of solid-state materials. Thus, the clarification of the interaction is required for understanding basic physical properties of solid-state materials and developing high-performance devices [1,2]. To clarify the interaction, it is important to understand the relation between the electronic state and the excitation of a particular lattice vibration (phonon).

Mode-selective phonon excitation (MSPE) is one of the useful methods for clarification of the relation between electronic state and the excitation of a particular phonon mode. Especially, a mid-infrared pulse laser is strong tool for MSPE. By irradiating a mid-infrared pulse laser tuned absorption wavelength of a specific phonon, the direct excitation of a specific phonon mode is available [3,4]. We have developed a technique which can directly observe excitation condition of particular phonon mode by using anti-Stokes Raman scattering spectroscopy (Fig. 1) [3]. By using the technique, the MSPE by MIR-FEL has been directly demonstrated with the sample material of 6H-SiC (Fig. 2) [3]. Then, we have started development of phonon dynamics measurement system by MIR-FEL and pico-second laser to investigate the differences of phonon property between selective excitation and thermal (non-selective) excitation.

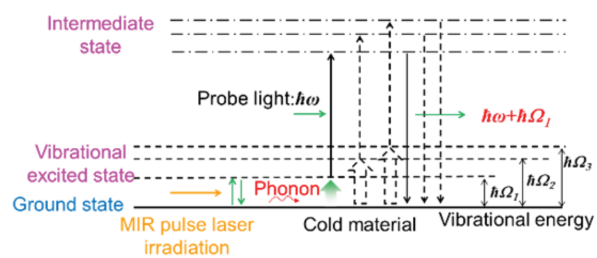

Figure 1: Schematic of the principle of anti-Stokes Raman scattering by MIR laser irradiation with cold material [3].

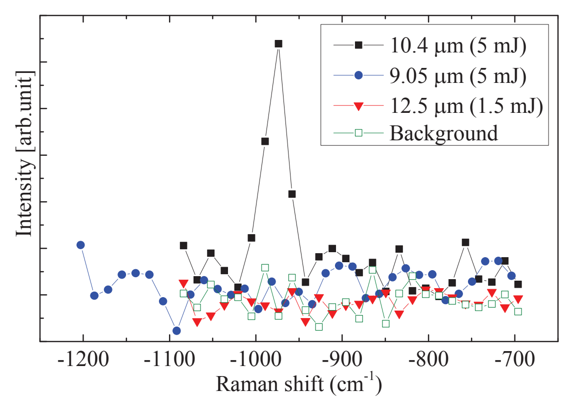

Figure 2: Anti-Stokes Raman scattering spectra with MIR-FEL and nano-second laser at 14 K [3].

PHONON DYNAMICS MEASUREMENT SYSTEM

Figure 3 shows the schematic diagram of developed phonon dynamics measurement system. In this system, the second harmonic of pico-second Nd-YVO_4 laser (probe light) [5] and MIR-FEL (pump light) are simultaneously irradiated. The wavelength and pulse

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width of the pico-second laser were 1064 nm, 7.5 ps, respectively. To pick up the near-infrared light, mirror was installed in front of half-wave plate before SHG crystal [see Fig. 2 of Ref. 5], and transported around 15 m to the user room. A beam expander was installed to parallelize the laser beam. The transported laser light was injected to BBO crystal for second harmonic generation. A bandpass filter was used to cut light from the fundamental wavelength (1064 nm). A quartz lens focused the laser on the sample surface. The pico-second laser is synchronized to the RF signal which was used for electron beam acceleration for MIR-FEL by using a piezo stage in the pico-second laser oscillator. The timing delay of MIR-FEL pulse against the pico-second laser can be adjusted by using a mechanical phase shifter installed in the RF signal line of driver linac of the MIR-FEL and an optical delay was also used. The emitted light from the sample was collimated by a quartz lens and focused on the entrance slit of the monochromator (Triax 190, Horiba Scientific). Notch filters (NF533-17, thorlabs: central wavelength of 532-nm and blocking bandwidth of 17-nm) were installed to supress the background photons caused by Rayleigh scattering on the sample. The spectrum of the emitted anti-Stokes Raman scattering light was measured by using a monochromator and a photomultiplier with a gating system (R3896 and C1392, Hamamatsu Photonics) under the photon counting condition. The gating timing of photomultiplier was synchronized to the oscillation timing of MIR-FEL. The output current signal from the photomultiplier tube was measured by a digital oscilloscope.

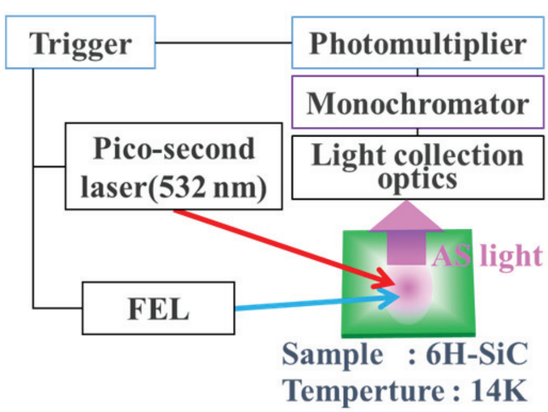

Figure 3: Schematic of the measurement system for anti-Stokes Raman scattering.

Table 1: Phonon Modes of 6H-SiC [6]

Wavenumber (Wavelength)	Infrared active	Raman active
965 cm^{-1} (10.4 μm)	○	○
797 cm^{-1} (12.5 μm)	○	×
787 cm^{-1} (12.7 μm)	×	○
767 cm^{-1} (13.0 μm)	×	○

TEST MEASUREMENT

Test experiments have been performed to evaluate the performance of the developed system and investigate the phonon dynamic of 6H-SiC which is the same sample with reference [3] and was a commercially available SiC (6H-semi-insulator type SiC, Xlamen Powerway Advanced Material Co., LTD:SiC) with dimension of 15 mm × 15 mm × 0.33 mm and the crystal orientation of (0001). First, the sample was cooled to 14 K using a closed-cycle Helium refrigerator, and thermal phonon excitation was suppressed. The known phonon modes of this sample around 10 μm are listed in Table 1 [6]. The FEL wavelength was tuned to 10.4 μm and the delay between the pico-second laser and MIR-FEL was varied to find the condition of simultaneous irradiation of two lasers. Next, the anti-Stokes scattering light intensity depending on the delay was measured to know the temporal resolution of the developed system. Moreover, under the simultaneous irradiation condition, the wavelength of MIR-FEL was varied to check the dependence on MIR-FEL wavelength.

RESULTS AND DISCUSSION

Timing Scan

To investigate the temporal resolution of the developed system, the MIR-FEL wavelength was tuned to 10.4 μm. Figure 4 shows the dependence of anti-Stokes Raman scattering intensity on the delay of the MIR-FEL. Horizontal axis indicates the delay time of the probe light (pico-second laser) against the pump light (MIR-FEL). At the optimum timing, we observed a strong anti-Stokes Raman scattering light at the Raman shift of -975 cm^{-1} as obviously shown in Fig. 4. As is shown in Fig. 4 the width of the delay time was around 20 ps in FWHM and the temporal resolution was evaluated around 10 ps. It would be worth to note that there is very weak tail in the positive delay side in Fig. 4. The origin of this tail is possibly decayed lattice vibration.

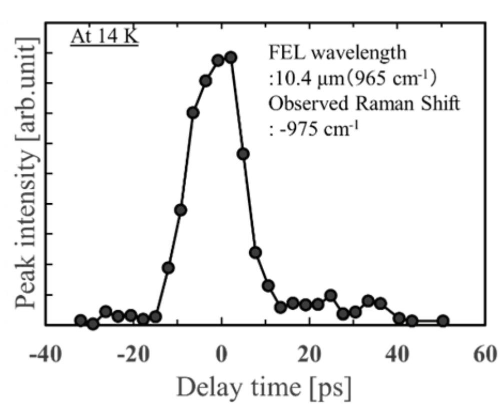

Figure 4: Dependence of anti-Stokes light intensity on delay time pico-second laser against MIR-FEL.

Since the predicted lifetime of the observed phonon excitation mode in 6H-SiC is about 2 ps at 20 K [use

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Eq. 5 of Ref. 7], the developed measurement system could not extracted any detail information of the lifetime by this test experiment. The developed system, however, can be available to measure the samples of longer lifetime, i.e. hydrogenated amorphous silicon, whose phonon lifetime are several tens pico-second [8].

FEL Wavelength Dependence

Figure 5 shows the measured anti-Stokes Raman scattering spectra at 14 K. When the wavelength of MIR-FEL was tuned to 10.4 µm, a peak emerged at 975 cm^{-1}. Comparing to the previous system which used a nano-second laser, S/N ratio was improved from 4:1 to 22:1. But when the wavelength of MIR-FEL was tuned to 8.95 µm and 9.60µm, the peak was observed in wavelength that does not correspond to absorption energy of lattice vibration of 6H-SiC. On the other hand, the sample has infrared active and Raman inactive phonon mode at 12.5µm, but anti-Stokes light peak was not observed in the case of 12.5-µm FEL irradiation. Consequently, either excitation of a special vibrational mode or sum frequency generation was suspected as a cause. Figure 6 shows dependence of wavenumber of the peak in anti-Stokes Raman scattering spectra on FEL wavenumber. The peak wavenumber of the observed anti-Stokes Raman scattering light was almost linear to the wavenumber of MIR-FEL used as the pump laser. Further studies in experimental and theoretical studies are required to reveal the phonon excitation mechanism by a mid-infrared pulse laser.

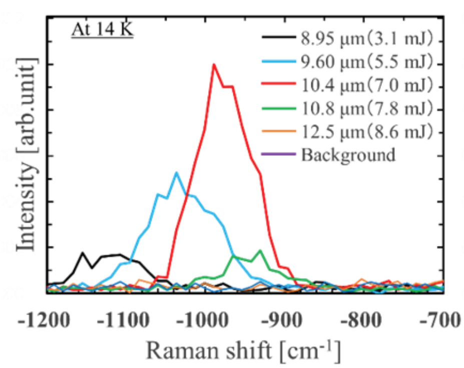

Figure 5: Anti-Stokes Raman scattering spectra with MIR -FEL and pico-second laser at 14K.

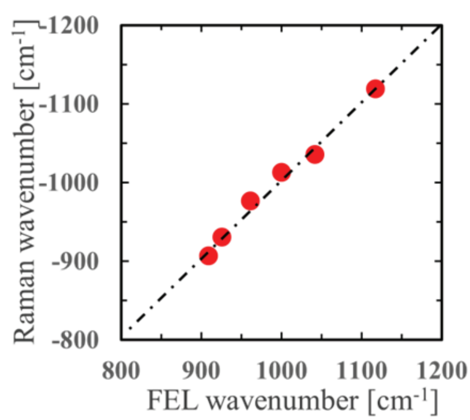

Figure 6: Dependence of the peak wavenumber in anti-Stokes light on FEL wavenumber.

CONCLUSION

We developed a phonon dynamics measurement system by MIR-FEL and pico-second laser to study the differences of phonon property between a selective excitation and a thermal (non-selective) excitation. The system was checked by using 6H-SiC which has been studied previous nano-second laser system. The test measurement showed that by using pico-second laser, S/N ratio was enhanced by 5 times in comparison to the nano-second laser.

The temporal resolution of the developed system was evaluated to be around 10 ps. Therefore the developed measurement would be available to the samples whose phonon lifetime are longer, i.e. hydrogenated amorphous silicon which has several tens pico-second of lifetime.

In addition, we have observed unexpected peaks in wavelength (8.95, 9.60 µm) during this study which do not correspond to any known phonon mode in 6H-SiC. The sample has infrared active and Raman inactive phonon mode at 12.5µm. But anti-Stokes light peak was not observed in the case of 12.5-µm FEL irradiation. Further study is required to reveal the phonon excitation mechanism by a mid-infrared pulse laser.

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