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Time-Resolved Optical Spectroscopy of a GaAs Single Crystal Irradiated by SR X-Ray Pulses

Yoshihito Tanaka[1, 2, a)], Maki Nagashima[1, 2)], Kenta Tanaka[1, 2)], Takayuki Hasegawa[1, 2)], Yoshimitsu Fukuyama[3)] and Nobuhiro Yasuda[3)]

[1]*Graduate School of Material Science, University of Hyogo, 3-2-1 Kouto, Kamigori, Ako, Hyogo 678-1297, Japan*
[2]*RIKEN SPring-8 Center, 1-1-1, Kouto, Sayo-cho, Sayo-gun, Hyogo 679-5148, Japan*
[3]*JASRI (Japan Synchrotron Radiation Research Institute), 1-1-1, Kouto, Sayo, Sayo-gun, Hyogo 679-5198, Japan*

[a)]Corresponding author: tanaka@sci.u-hyogo.ac.jp

Abstract. Brilliant synchrotron radiation (SR) has enabled us to investigate fast phenomena even in small samples. At the same instance, the property of sample may be affected by the SR due to the high brilliance. An SR X-ray pump and optical probe measurement has then been performed at a storage ring facility of SPring-8 to investigate the spectral change in the near infrared region of a GaAs single crystal irradiated by an intense X-ray pulsed SR beam with a photon energy of just above the K-edge of As. The transient transmission spectrum was monitored by using a broadband optical pulsed beam and a dispersive spectrometer. We obtained the spectral profile change with a response time of shorter than the pulse duration of SR. The spectrum shows the abrupt change in transmittance at around 880 nm corresponding to the band gap energy of GaAs, and the fringe on the spectrum originated from the interference at the sample with a thickness of about 10 μm. The band gap energy and the fringe shift to the opposite direction to each other, the behavior of which is explained to be due to the band gap shrinkage accompanied with the decrease of refraction index originated from the free carriers. The transient band gap shrinkage suggests the nonthermal lattice distortion induced by SR irradiation.

INTRODUCTION

Recent progress of brilliant SR sources has enabled us to perform quick X-ray measurement with high sensitivity. Furthermore, the pulsed nature has allowed the time-resolved measurements for fast phenomena even in small samples. At the same instance, the effect of X-ray irradiation to the sample property may be no longer negligible due to the high brilliance. In fact, the temporal changes in the optical transmission of GaAs and Si_3N_4 have already been utilized for the crosscorrelation to monitor the timing between an optical laser pulse and an X-ray free electron laser pulse [1-3]. Even in experiments at a storage ring, optical and electronic properties of semiconductors sensitive to photoexcitation may change by X-ray SR irradiation for probe during the measurement. This means that when we measure optical and electric parameters of a target semiconductor sample by using an intense SR beam, the measured value may be different from the original one without SR irradiation.

When a semiconductor is irradiated by a synchrotron X-ray beam, the inner shell electrons are initially excited, and a lot of electrons then occupy the conduction band through the successive Auger processes. The electrons finally go back to the original state in the valence band through the recombination process along with emitting photons and phonons. As the process occurs quickly, *i.e.* excitation and recombination processes occur in femto- to nanoseconds, X-ray SR pump and optical probe method is used to investigate the optical property during the process. As the absorption spectral profile around the band gap energy involves the information on electronic states, we use the broadband pulsed laser for probe in the experiment. In this paper, we report the transient transmission spectrum around the band gap energy of GaAs, when the sample was irradiated by the X-ray pulses with high power density at the storage ring facility of SPring-8, and discuss the influence on the sample properties, such as optical and electronic and lattice states.

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EXPERIMENTAL

Figure 1(a) shows a schematic of the experimental setup for SR pump and NIR (Near Infrared) probe experiment at the SPring-8 storage ring facility. The experiment was performed at BL40XU, the ID (insertion device) of which is a helical undulator producing circular polarized X-ray beam with less high-harmonics components of X-ray photons at around the beam axis. The beam with a flux of about 10^{15} photons/s was guided to the experimental hutch through curved mirrors for two-dimensional focusing, without a crystal monochromator. In the experimental hutch, a non-doped GaAs single crystal plate was irradiated by the X-ray beam with an incidence angle of 45 degrees and a size of ~40 μm (V) ×300 μm (H). The thickness of the GaAs plate was designed to be 10 μm, in order to measure the absorption spectrum with the interference pattern in the NIR region. The sample was made by way of the process that a GaAs wafer was attached on a sapphire plate, and was ground down to be around 10 μm in thickness, and the GaAs portion was then peeled by heating. The GaAs plate with a size of 3 mm by 3 mm, was attached on a glass capillary with epoxy resin adhesive. The beamline is equipped with a mode-locked Ti:sapphire laser system composed of a femtosecond laser oscillator and a regenerative amplifier. The output pulse energy, wavelength, pulse width, and repetition rate are 1 mJ/pulse, 800 nm, 130 fs, 1 kHz, respectively. The laser beam was guided to a sapphire plate to generate a broadband light pulse. The effective spectral region is 720-960 nm, which can cover the region of the band gap of GaAs. The broadband pulses illuminated the GaAs sample plate through a lens with an incidence angle of 45 degrees (perpendicular to the X-ray beam), and the transmitted beam was guided to an optical fiber connected to a dispersive spectrometer.

The X-ray photon energy was tuned by the ID gap in order to excite the K-shell electrons of As as well as that of Ga of the sample. Figure 1(b) shows the transmittance of the helical undulator beam in the range of K-edges of Ga and As. The edges seem smooth because the fractional bandwidth of X-ray beam at a beamline of BL40XU is around 2 %. The photon energy of the fundamental peak of undulator spectrum was tuned to be around 12.1 keV.

The filling pattern of the storage ring was so-called hybrid mode, which includes one isolated single bunch with high bunch current together with the multi-bunch section in a third of buckets. An X-ray pulse chopper with a repetition rate of 1 kHz was used to pick up the X-ray pulse from the single bunch. As the Ti:sapphire laser system is synchronized to the master clock of the storage ring, the obtained broadband pulses are also synchronized to the X-ray synchrotron pulses. An X-ray pump and NIR broadband light probe measurement was then performed by controlling the time delay between these pulses with a continuous phase shifter of an RF signal for laser trigger.

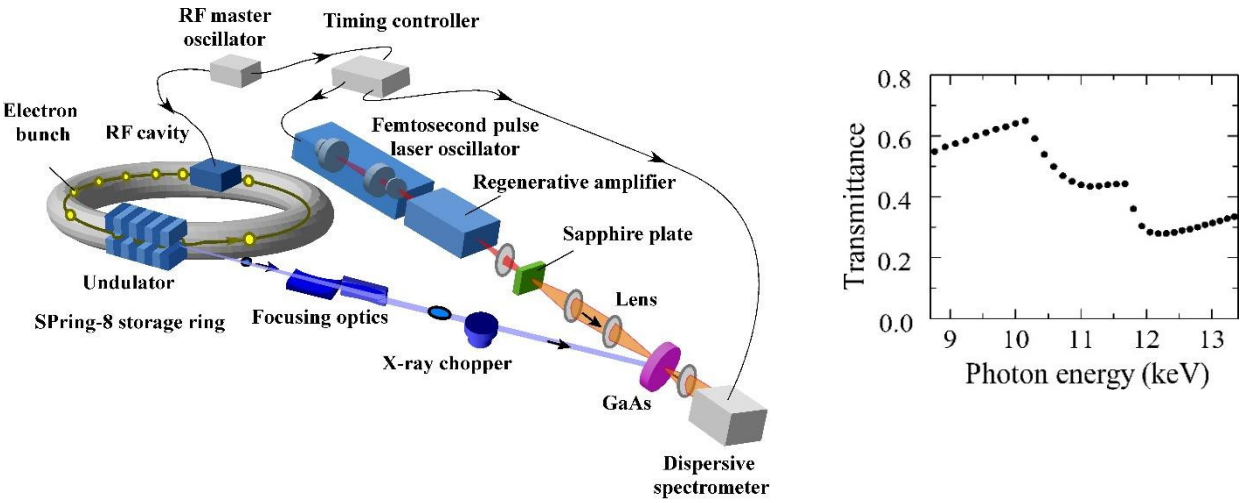

FIGURE 1. (a) A schematic of experimental setup for time-resolved NIR spectroscopy of GaAs irradiated by SR X-ray pulses. (b) Transmittance of the 40XU undulator beam for a GaAs sample at around K-edges of Ga and As.

RESULTS AND DISCUSSION

Figure 2 shows the NIR transmission spectra of the GaAs plate. The large difference in transmittance is seen between the longer and the shorter wavelength regions than around 875 nm corresponding to the band gap energy. The fringe pattern seen in the longer wavelength region is originated from the interference between the sample surfaces. Thus the period is related to the thickness and the refractive index. The X-ray pulse energy density for pump in Fig. 2 is 0.6 mJ/pulse/cm^2, where the 3×10^7 photons/pulse is concentrated to the area of 39 μm (V) × 340 μm (H). The thick red and dashed blue curves are the spectra at the time delays of 200 ps and -200 ps, respectively. The threshold wavelength where the transmittance becomes drastically larger, shifts to the direction of longer wavelength. The inset shows the time-resolved transmittance at 880 nm indicated by a dashed line in Fig. 2. The transmittance drops quickly at time-zero and relaxes with a time constant of several nanoseconds.

From the result that the spectral profile "shifts" to the longer wavelength, it is found that the band gap shrinks by X-ray excitation. Then the fast speed in the initial change appeared in the inset is thus reflected by the band gap change. It is known that the band gap shrinkage is strongly correlated with the lattice expansion. At the time scale, lattice expansion involves both thermal and non-thermal processes. If a major contribution to the lattice expansion is assumed to be attributed to thermal effect, the change in temperature is estimated to be about 2 K, as the coefficient of band gap change for unit temperature is -0.46 meV/K (0.3 nm/K at 880 nm) for GaAs. The thermal effect should also be accompanied by the change in the thickness and the refractive index. In the case of GaAs, the peak or dip of the fringe shifts to the longer wavelength by ~0.6 nm for temperature increase by 2 K. However, the fringe shift cannot be seen during a few ns as in Fig. 2. Thus the band gap shift is dominated by nonthermal processes.

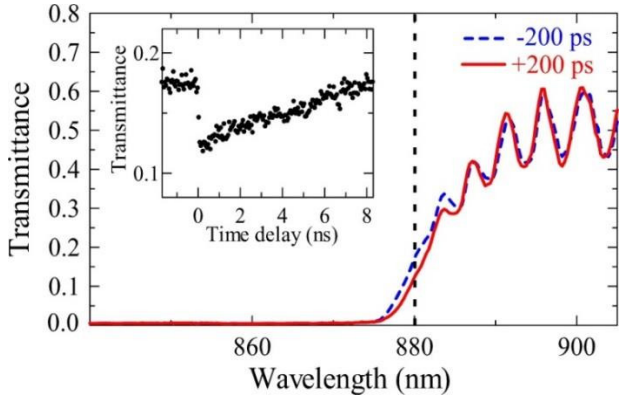

FIGURE 2. Time-resolved transmission spectra in the NIR region of a GaAs plate irradiated by SR X-ray pulses with Δt (time delay) = -200 ps (blue) and +200 ps (red). Inset: Time-delay dependence of transmittance at 880 nm.

In order to see the fringe shift clearly, the pump pulse energy density was increased. Figure 3(a) shows the time-resolved transmission spectra for a pump pulse energy density of 2.9 mJ/pulse/cm^2, which is five times larger than in Fig. 2. It is clearly seen that the fringe on the transmittance at a time delay of 100 ps shifts to the shorter wavelength direction, which is opposite to the case in temperature rise. The fringe shift is explained to be due to the decrease of refraction index originated from the free electron carriers [3]. It is concluded that X-ray excitation of GaAs creates dense free electron carriers and induces the band gap shrinkage.

Figure 3(b) shows the time evolution of the threshold photon energy, E_{th}, which is corresponding to the band gap energy, in order to estimate the speed of the change. The threshold energy, E_{th}, is determined from Fig. 3(a), through the procedure as follows: The axes scales in Fig. 3(a) are converted to the products of absorbance squared and photon energy squared, and the corresponding photon energy for vertical and horizontal axes, respectively. The intersection point between the linear function fitted around the drastic change and the horizontal axis at zero absorbance then determines E_{th}. The constant shift of E_{th} at >50 ps in Fig. 3(b) indicates that the recombination time is much longer than a few hundreds of ps. The graph is thus fitted by a sigmoid function (dashed curve). The derivative is also drawn with a solid red curve on the same time-axis, the FWHM of which is (26 ± 10) ps. As the time resolution of the measurement system is a few tens of ps, which is determined by the pulse width of SR, it is found that the phenomenon is much faster than the few tens of picoseconds. The origin of the fast response to be

considered is the lattice temperature rise through electron-lattice thermalization, the speed of which is ps order in GaAs [4].

It is also suggested that the quick band gap change may be due to nonthermal lattice distortion induced by photo-irradiation. In GaAs, the lattice space expands through the nonthermal effect, resulting in band gap shrinkage. The assumption may also be supported by the carrier density in the conduction band estimated from the absorption efficiency of X-ray photons. Table 1 shows the photo-excited carrier densities estimated for various pump probe experiments. The carrier density estimated in the present experiment is comparable to that in the optical pump X-ray probe experiment for the observation of nonthermal effect in GaAs [5].

In conclusion, brilliant X-ray irradiation of GaAs brings high-density carrier generation and the quick band gap shrinkage. The fast shrinkage of band gap may be accompanied with nonthermal lattice distortion within the X-ray pulse duration. The effect impinging on the lattice spacing should be examined at X-ray diffraction experiments using brilliant SR X-ray beams.

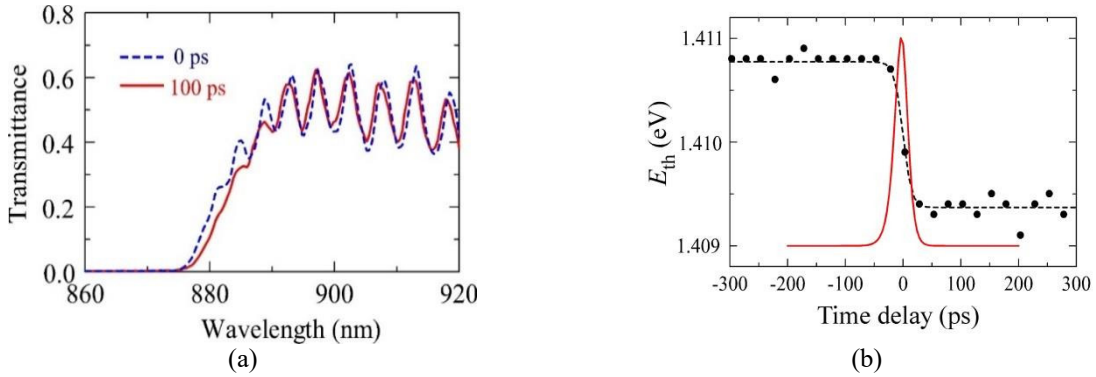

FIGURE 3. (a)Time-resolved transmission spectra in the NIR region of a GaAs plate irradiated by SR X-rays with a pulse energy density of 2.9 mJ/pulse/cm^2. (b) Change in the threshold photon energy, E_{th}, at around a time delay of 0 ps. The data are fitted by a sigmoid function (dashed curve), the derivative of which is described on the graph in red.

TABLE 1. Photo-excited carrier densities estimated form the absorption efficiency of optical or X-ray photons for various pump probe experiments

Pump and probe experiments at SR facilities	Carrier density	Reference
X-ray pump & NIR probe at APS	5.0×10^{19} electrons/cm^3	A. Durbin et al. [1]
X-ray pump & NIR probe at SPring-8	3.9×10^{18} electrons/cm^3	This paper
NIR pump & X-ray probe at SPring-8	2.4×10^{18} electrons/cm^3	Y. Tanaka et al. [5]

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