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Ultrafast optical Kerr effect of excitons weakly confined in GaAs thin films

Atsushi Kanno[1,2,*], Redouane Katouf[1,2], Osamu Kojima[1,2,3], Junko Ishi-Hayase[1,2], Masahiro Tsuchiya[1,2], and Toshiro Isu[1,2,4]

[1] National Institute of Information and Communications Technology, 4-2-1 Nukui-kitamachi, Koganei, Tokyo 184-8795, Japan
[2] CREST, Japan Science and Technology Agency, Japan
[3] Department of Electrical and Electronics Engineering, Kobe University, 1-1 Rokkodai, Nada, Kobe 657-8501, Japan
[4] Instiitute of Technology and Science, The University of Tokushima, 2-1, Minamijosanjima-cho, Tokushima 770-8506, Japan

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* Corresponding author: e-mail kanno@nict.go.jp, Phone: +81-42-327-6876, Fax: +81-42-327-5328

We investigate ultrafast response of optical Kerr effect (OKE) induced by excitons weakly confined in GaAs thin films. The observed decay time of optical response agrees approximately with incident pulse duration. At 100-fs pulse irradiation the response time is estimated to be 170 fs, though the exciton lifetime is about 10 ns. It is considered that the ultrafast response of OKE is caused by not population relaxation but orientation relaxation of excitons.

1 Introduction The optical characteristic of an exciton in quantum structures has been intense research target for ultrafast and high nonlinear optical devices. When the center-of-mass motion of exciton is confined in a thin film, where the thickness is larger than exciton Bohr radius, an ultrafast response and high nonlinearity was reported theoretically under the nonlocality-induced double resonance in energy and size (NIDORES) condition [1]. Under ps-pulse irradiation, such interesting features were observed by transient grating measurements [2,3]. Under fs-pulse irradiation, the optical response showed not only sub-ps-order decay constant but also long-decay component by the transient grating and degenerate four-wave-mixing measurements [4]. In time-resolved photoluminescence (PL) measurements, on the other hand, the PL time constant was measured to be about 10 ns [5]. This indicates that the weakly confined exciton is long-lived in thin film. The long lifetime of exciton population is one of disadvantageous properties for applying all-optical ultrafast device. However, the previous report of optical Kerr effect (OKE) measurement in the excitons showed high and fast response under ps-laser irradiation [6]. The OKE is suitable effect for optical polarization switch because the phenomenon causes the change of the refractive index. In addition, the OKE measurement is adequate technique for investigation of nonlinear properties of weakly confined exciton because the OKE is based on third-order nonlinear effect. In this paper, we observed time-resolved OKE profiles of weakly confined exciton with various duration laser pulses. The fast component of observed decay profile was near to the incident pulse width. The response time of the OKE is estimated to be 170 fs under 100-fs pulse irradiation. This indicates that ultrafast optical response appears via nonlinear optical process such as the OKE, though the exciton population lifetime is very long.

2 Experimental setting The sample consisted of three double-hetero-structure layers, which were made of GaAs active layer (110 nm thick) sandwiched by $Al_{0.3}Ga_{0.7}As$ layers (5.7 nm thick) grown by molecular beam epitaxy on (001) GaAs wafer. Under the NIDORES condition, it was reported that the high nonlinearity and fast response appeared at the GaAs thickness of 110 nm [2]. An excitation laser was a mode-locked Ti:sapphire laser pumped by Nd:YVO_4 green laser. The sample was set in cryostat and was kept at 4 K in all measurements. Time-resolved

PL measurements were performed using a streak camera with a single monochromator under normal incident condition. For measurement of a long decay, we used the slow repetition laser with 4 MHz. In order to avoid detecting the scattering of incident laser light, the angle of the polarizer of laser was set to be perpendicular, so-called cross-Nicol configuration, to the one of the analyzer of PL detection.

Time-resolved OKE measurements were performed using typical pump-probe measurement system with polarization detection configuration (Fig. 1). Pump and probe pulses were linearly polarized, and the polarization angle between them was set to be 45 °. The angle of the polarizer and analyzer was set to perpendicular for detection of polarization change. For obtaining high signal-to-noise ratio and rapid measurement, a fastscan technique was used in this experiment [7, 8]. The slow repetition rate shaker, which was driven at 4 Hz, was inserted in pump-laser line, and was used as optical delay between pump and probe pulses. The maximum length of optical pass difference corresponded to ±75 ps. The mechanical stage, which was inserted to the probe line, was used for fine adjustment of optical path difference and delay time calibration. The reflected probe pulse from sample was detected by fast photodetector through the analyzer, and accumulated by sampling digital oscilloscope. Since a signal was detected with shaker position synchronously, the information of delay time between pump and probe pulses was recorded at the same time. It was confirmed that the signal measured by the fastscan technique was equal to the one measured by the traditional pump-probe technique. The duration and repetition rate of the excitation laser were 100 fs and 80 MHz, respectively. The laser linewidth was varied by a spectral filter unit using a slit and grating pairs. The variation range was from 1 to 25 meV, which corresponded to several ps to about 100 fs pulse.

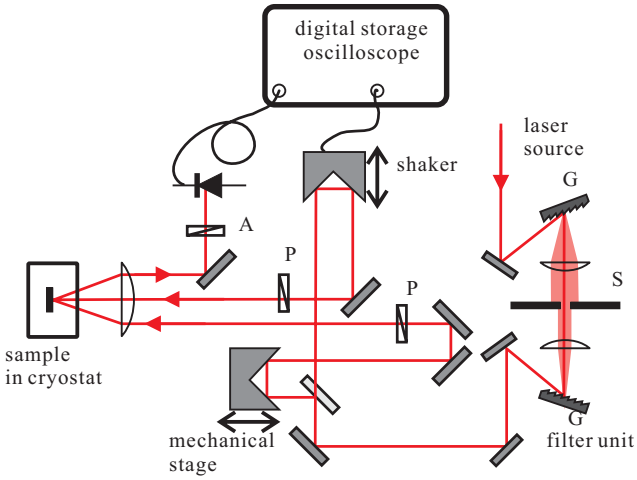

Figure 1 Experimental setup of time-resolved OKE measurements. G, S, P and A indicate grating, slit, polarizer and analyzer, respectively. In traditional pump-probe measurement, the mechanical optical chopper (not shown in the figure) was inserted to the pump laser line.

3 Results and discussion The PL was taken under non-resonant (high energy) excitation with a low power enough to avoid a multi-exciton effect. Reflection spectrum was measured at Brewster angle with halogen light. Figure 2 clearly shows good agreement with peaks of the PL and edges of the reflection spectra. These spectra indicate that the weakly confined exciton surely exists in the thin film. In addition, the figure clearly shows that the excitation light generated only free excitons in thin film because the other exciton levels (such as bound exciton states) cannot be observed. Figure 3 shows the time-resolved PL profile under resonant excitation condition. The time constant is estimated to be 14 ns. It is considered that high quality sample we fabricated causes such a long lifetime because PL peaks originating from impurity centers are hardly observed.

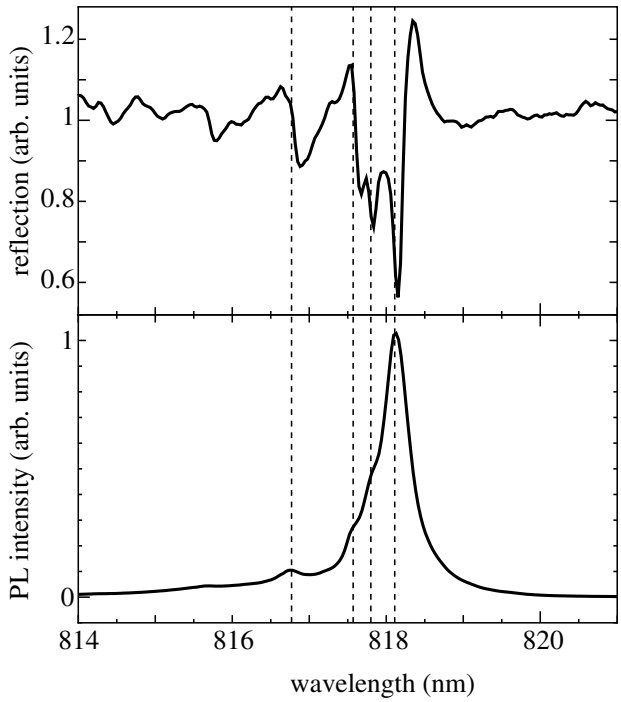

Figure 2 Reflection (upper) and PL (bottom) spectra at 4 K. Dashed lines are guidelines for eyes.

Figure 4 shows the result of time-resolved OKE profiles of weakly confined excitons with various pulse spectral widths. The variation of spectral widths corresponds to the change of the incident pulse duration. Average powers

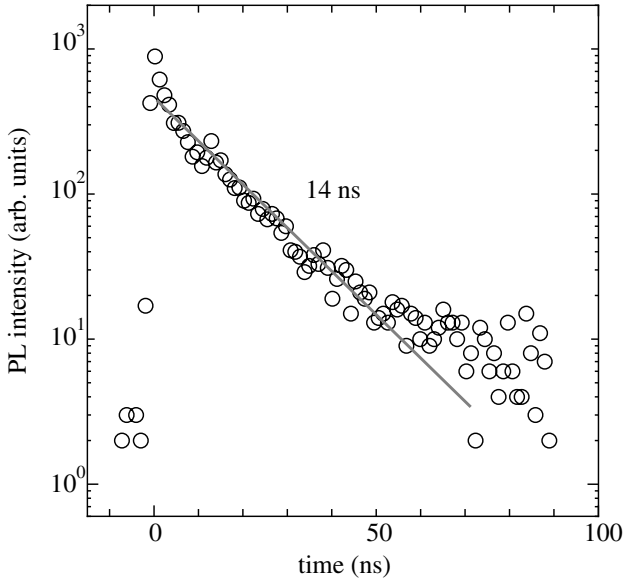

Figure 3 Time-resolved PL profile detected at PL peak energy (818 nm) with streak camera under resonant excitation condition. Solid thick line is the guideline for eyes.

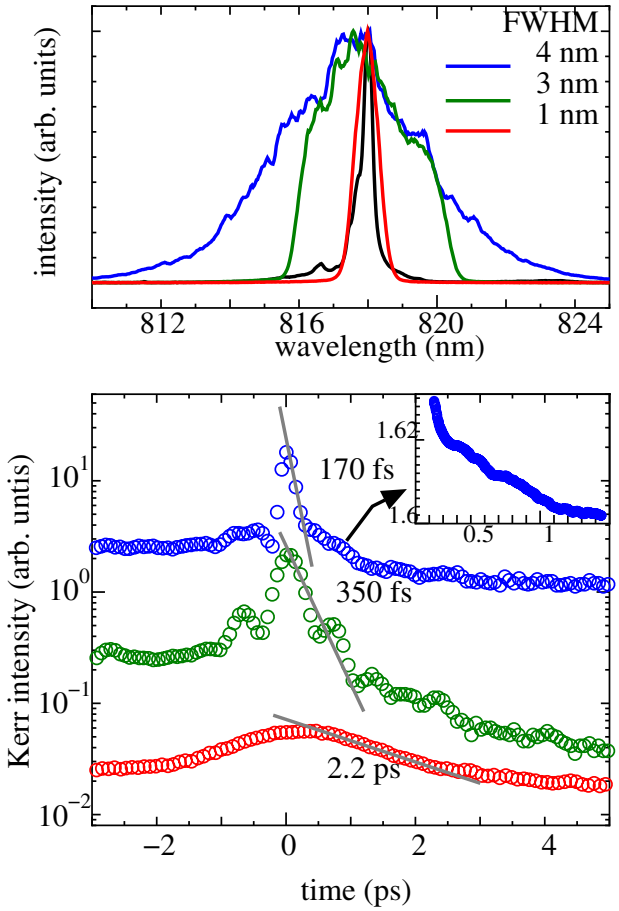

Figure 4 Upper figure shows PL (black line) and irradiated laser spectra at 4 K. Bottom figure shows time-resolved OKE signal at the same temperature. Solid gray lines are fitting lines of observed decay signals. The color of symbols corresponds to linewidth of irradiation laser pulse in the upper figure. Inset: magnified view at shortest pulse duration in linear plot. Weak and short-period oscillatory signal is observed.

of incident pump pulses were set to be the same of 100 μW, and those of the probe pulse were set to be a tenth of the pump pulses. When the pulse width decreased, decay time of the Kerr signal decreased. The decay time was estimated by single exponential fitting of the observed signal. Typical decay time was estimated to be 170 fs under fs-laser pulse irradiation. This indicates that the OKE response has ultrafast component as fast as the incident pulse duration. The observed decay time was 1.6 ps under ps-laser pulse irradiation. The behavior of the observed OKE profile is comparable with the reported one [6]. Note that the exciton lifetime is very long about 10 ns described above. It is considered that relaxation mechanism is different from the one of the exciton population relaxation. The orientation relaxation of excitons should be dominant relaxation process in OKE response. In addition, the OKE signal at the delay time of several ps in the figure is smaller than that at the negative delay time. It is considered that the effect due to generation of long lifetime excitons under resonant excitation condition causes the change of the observed signal intensity. Note that oscillatory structures were observed when the incident pulse linewidth was broader. Although oscillatory signal was very weak for the broadest pulse linewidth irradiation, shorter period structure was observed, as shown in the inset of Fig. 4. This is caused by an excitation of multi energy levels of the weakly confined exciton. The effect of the excitation of multi energy levels was reported in degenerate four-wave-mixing measurement in the same sample [9].

Pump-laser power dependence of the peak intensity of observed OKE signal was measured (Fig. 5). The average power of probe pulse was set to be 1 μW. Above 200 μW, the relationship between the input power and the peak intensity strongly disagrees with the I^2 line. The high excitation power effect, such as the generation of multi-exciton, causes the behavior of the input power dependence. The critical power (100 μW) agrees with the reported one measured by the other nonlinear measurements [4, 10].

4 Conclusion We investigate the ultrafast OKE response induced by weakly confined exciton in GaAs thin films under NIDORES condition. Although the exciton lifetime is very long about 10 ns, the decay time of OKE response is near to the incident pulse duration, and the observed value achieves to be 170 fs at the pulse duration of 100 fs. This indicates that the relaxation mechanism of Kerr effect is not exciton population but orientation relax-

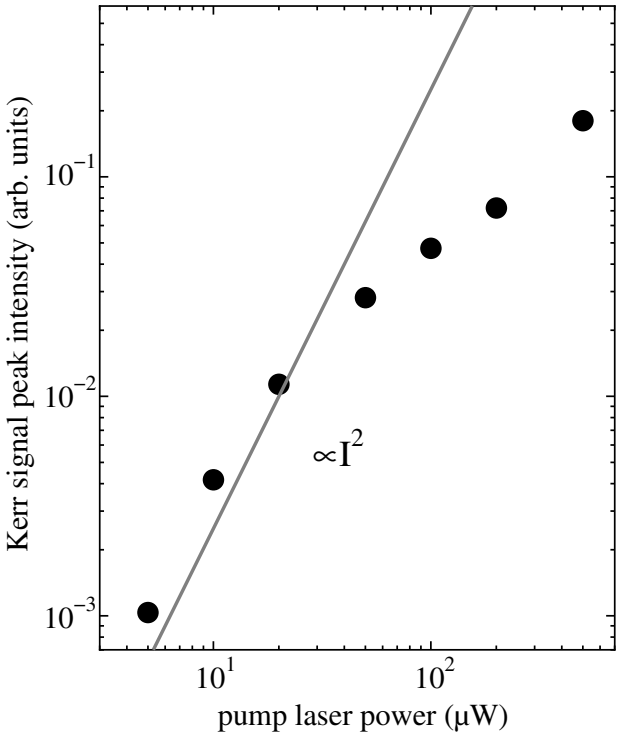

Figure 5 Input pump laser power dependence of OKE signal at zero ps at 4 K. The probe power is set to be 1 μW. Solid line is the guideline of proportional to I^2.

ation. The ultrafast response of weakly confined exciton has possible candidate for new optical devices.

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