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Decay of orientational grating of weakly confined excitons in GaAs thin films

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Abstract

We report the dynamical properties of the exciton orientation in GaAs thin films using the orientational grating (OG) technique. From the results of excitation-power dependence of OG signal, we confirmed that the OG signal comes from the optical nonlinearity of weakly confined excitons. In addition, the OG-decay time decreases with an increase of excitation power due to exciton–exciton interaction, and the shortest decay time is below 1 ps. Our results may imply the potential application of optical nonlinearity of weakly confined exciton to ultrafast switching devices operating at 1 Tbit/s.

Keywords: Weakly confined exciton; Nonlocal response; Thin film; Optical nonlinear response; Transient grating

1. Introduction

Recently, ultrafast optical switching devices have been increasingly needed for information technology. For optical switching devices such as optical Kerr shutter, the clarification of the in-plane rotation process of excitons, leading to the polarization control, is an important factor. Moreover, high optical nonlinearity also is needed for ultrafast switching devices. It has been reported that the weakly confined excitons in GaAs thin films with a specific film thickness have large optical nonlinearity due to the nonlocality-induced-double resonance in energy and size (NIDORES) [1,2], where the weakly confined excitons are characterized by nonlocal-response theory [3,4]. In addition, we reported that the population decay and the dephasing profiles of the weakly confined excitons depend on the spectral width of excitation pulse [5,6]. Then, if the orientation-decay profile of the weakly confined excitons under NIDORES condition is clarified, the exciton optical nonlinearity may lead to the realization of ultrafast switches based on the Kerr rotation. In the present work, we have investigated the orientation decay of weakly confined excitons in GaAs thin films by an orientational grating (OG) technique. We confirmed that the OG signal comes from the exciton optical nonlinearity from the excitation-power dependence. The possibility of ultrafast devices using excitons under the NIDORES condition is discussed from an aspect of the excitation-power dependence.

2. Experiment

A sample used in the present work is double heterostructure (DH) thin film with three periods of GaAs(110 nm)/$Al_{0.3}Ga_{0.7}As$(5 nm) on a (0 0 1) GaAs substrate grown by molecular beam epitaxy. The $Al_{0.3}Ga_{0.7}As$ barrier layer has enough thickness to confine the excitons in the GaAs thin films. In the case of film thickness of about 110 nm, the enhancement of optical nonlinearity of

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the $n=2$ confined exciton due to the NIDORES effect has been reported [1,2]. The decay profiles of exciton orientation were measured by an OG technique at 5 K in the backward direction of $k_1-k_2+k_3$. The light source was a mode-locked Ti:sapphire pulse laser with a 76-MHz repetition and a pulse width of 1.5 ps. The excitation energy was 1.5158 eV, which is the $n=2$ confined exciton energy with maximum optical nonlinearity in the sample. The excitation power was changed from 60 to 300 μW. The intensity ratio of the power of the excitation and probe pulses is 10:1. The polarizations of k_1 and k_2, which are the excitation pulses (formation of OG), were crosspolarized. The OG was produced by interference of two orthogonally polarized light fields [7]. The coherent superposition of the two crosspolarized incident pulses from different directions (k_1 and k_2) is spatially modulated in the direction of the resultant vector of electric fields, which forms the OG. Moreover, we measured a degenerate four-wave-mixing (DFWM) signal for reference. In the DFWM measurement, the two pulses (k_1 and k_2) were copolarized and a power of 100 μW corresponding to an excitation density of 12 nJ/cm^2. In both of the OG and DFWM measurements, the diffracted pulse was detected by the photodetector through a polarizer with the same polarization of probe pulses; k_3 and k_2 in OG and DFWM, respectively. For the detection, we did not employ the usual lock-in detection system but a fast-scan system using a shaker with frequency 20 Hz for improvement of the signal-to-noise ratio.

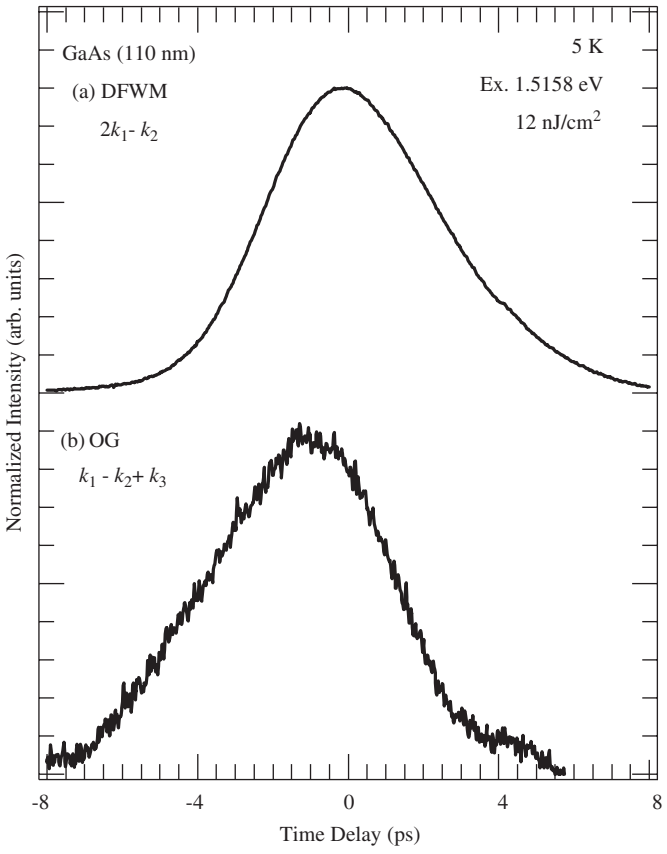

Fig. 1. (a) DFWM and (b) OG signals measured under exciton resonant condition.

3. Results and discussion

Figs. 1(a) and (b) show the DFWM and OG signals at an excitation power of 100 μW, respectively. It was reported that the optical nonlinearity of the exciton in the present sample shows cubic dependence on excitation power up to 12 nJ/cm^2 [1]. The decay time of the DFWM signal is 1.3 ps, which results from the dephasing of weakly confined exciton [8]. The decay time of the OG signal is 3.5 ps. The decay times were evaluated by a single exponential function form. We performed a fitting for the signals after 4 ps in order to eliminate the contribution of the temporal shape of excitation pulse. Schultheis et al. [9] regarded the decay time of the OG signal as the dephasing time. However, in our measurement, the decay time of the OG signal is different from the dephasing time. We reported a decay time of the population of over 100 ps [5]. This means the exciton population remains after the dephasing in our sample. Hence, the excitonic electric fields will remain after exciton dephasing.

The excitation-power dependence of OG signal for confirmation of the origins of OG signal is depicted in Fig. 2. The signal intensity increases with an increase of

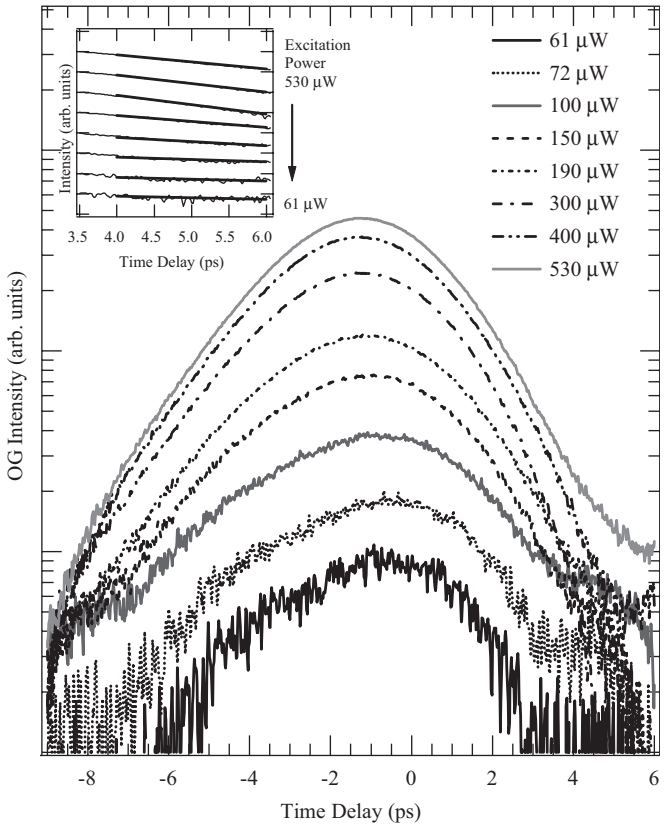

Fig. 2. Excitation-power dependence of the OG signal. The inset shows the fitting with a single exponential function form for each OG signal. The signals shown by thin curves were normalized at the maximum intensity, and the thick lines indicate the fitting results.

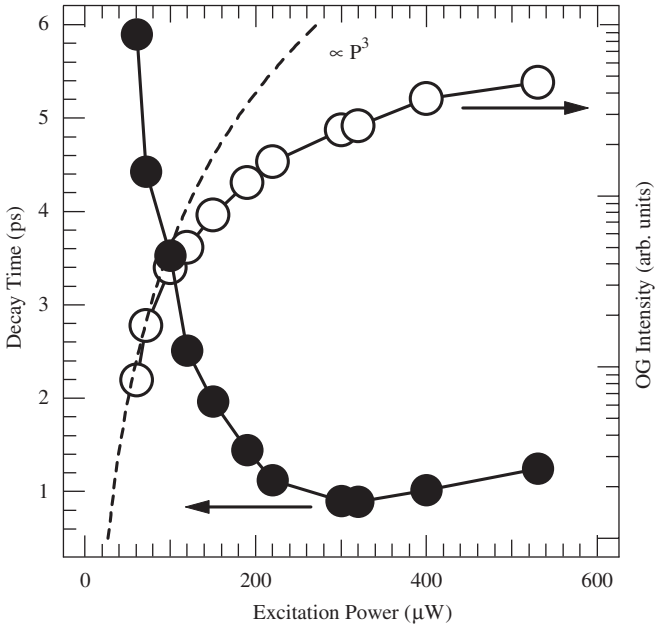

Fig. 3. The signal intensity (○) and decay time (●) plotted as a function of excitation power. The dotted curve indicates the cubic dependence of the intensity on excitation power.

excitation power. The signal-decay time obviously varies with the excitation power. For evaluation of the decay time of each OG signal, we performed a fitting with a single exponential function form in the time region 4.0–6.0 ps for the normalized signal at maximum intensity around zero delay, as shown in the inset of Fig. 2. The vertical axis of the inset is also logarithmic scale. The thick lines indicate the fitting results. The decay time and the intensity of the OG signal are plotted as a function of excitation power in Fig. 3. The dotted curve demonstrates the cubic dependence of the signal intensity on excitation power. Since the OG intensity indicated by open circle shows the cubic dependence up to about 100 μW, the OG signal originates from the exciton optical nonlinearity. The decay time of the OG signal indicated by filled circle decreases with an increase of excitation power, and becomes almost a constant value over ~300 μW. The shortest decay time is about 0.8 ps, which may be limited by the pulse width, and the decrease of OG decay time is due to the exciton–exciton interaction. The value of the shortest decay time includes the ambiguity due to the consideration of pulse width because the values of the decay time were automatically obtained by the fitting. Our results imply that the decay of the exciton orientation can be controlled by the excitation power, which is an important factor for the realization of ultrafast switching devices.

4. Conclusion

We have investigated the exciton-orientation dynamics by an OG technique. The decay time of the OG signal was longer than that of the DFWM signal. From the excitation-power dependence of OG signal intensity, which shows a cubic dependence, it was confirmed that the OG signal comes from the optical nonlinearity of weakly confined excitons. Moreover, the OG-decay time decreases with an increase of excitation power because of exciton–exciton interaction, and the shortest decay time is about 0.8 ps. Our results may imply the potential application of optical nonlinearity of weakly confined exciton to ultrafast switching devices operating at 1 Tbit/s.

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