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Spectrally resolved nonlinear optical response of weakly confined excitons under femtosecond laser pulse excitation in GaAs thin films

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We have investigated the dynamical properties of the weakly confined excitons in GaAs thin films by a transient grating method (three-beam four-wave mixing) with the use of femtosecond-laser pulses. The time-domain signals under the exciton-excitation conditions show a nonlinear optical response with a rise time of 4 ps and a long decay time of 120 ps. Moreover, in the case of that the probe pulse is parallel polarized to the two excitation pulses, the ultrafast optical response comparable to the pulse width appears. These characteristics of the nonlinear optical response is different from the experimental results by the picosecond-laser-pulse excitation. The spectra of the transient grating signals at the various time delays show several peaks at the energies, which agree with the energies of excitons confined in the thin films. This fact demonstrates that the characteristics of the nonlinear optical response of the weakly confined excitons are drastically changed by the spectral width of the excitation laser-pulse.

1 Introduction Dynamical properties and optical nonlinearity of excitons in semiconductor bulk crystals, multiple quantum wells, and quantum dots have been extensively studied from the viewpoints of physics of ultrafast phenomena and application to ultrafast optical devices [1, 2]. Recently, in the weak confinement regime, it was reported that optical nonlinearity and fast response of excitons due to the interaction with the radiation field are remarkably enhanced by the double resonance in energy and size [3–5]. However, to our knowledge, many experiments for the nonlinear optical response of the weakly confined excitons have been performed by using laser pulses with a width of a few picoseconds and narrow spectrum width which resonantly excites a single exciton state.

If femtosecond-laser pulses with a broader spectral width are used to generate nonlinear optical response of weakly confined excitons, multiple exciton states which are energetically close each other are excited. Thus, we may reveal the effect of the spectral width of the excitation pulse on the nonlinear optical response of the weakly confined excitons by comparing with the results of picosecond-laser pulse-excitations. In the present work, we have investigated the nonlinear optical response of the weakly confined excitons in GaAs thin films by using a transient grating method with 160-fs pulse-duration for the first time. We have found that the transient grating signals of the GaAs thin films under the femtosecond-pulse-excitation conditions indicate a rise time of 4 ps and a decay time of 120 ps. In addition, the ultrafast optical response comparable to the pulse width is observed when all of the pulses of the excitation and probe are copolarized. The origin of the nonlinear optical response is discussed from an aspect of the spectra of the transient grating signals.

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2 Experiment The sample used in the present work is a double heterostructure (DH) with 3 periods of GaAs(110 nm)/$Al_{0.3}Ga_{0.7}As$(5 nm) on a (100) 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 film [3–5]. The transient grating method was performed at 5 K. The transient grating method is a three-beam four-wave mixing in the case that the time delay between the two excitation pulses is zero, and the diffracted signal as a function of the time delay between the excitation and probe pulses provides the information about the population decay time of the system. The laser source used was a mode-locked Ti:sapphire pulse laser with a pulse width of 160 fs. The laser photon energy was tuned at 1.517 eV, which is around the exciton energies in the present sample. The spectral width of the laser pulse was 20 meV, so that the multiple exciton states were excited. The excitation power was kept at 1.2 pJ/pulse for 100 μm diameter. Below this excitation power, the intensity of the transient grating signal shows a cubic dependence on the excitation laser power (not shown here). Figure 1 shows the schematical experimental setup. The pulse train is split into three parts that are focused to a single spot on the sample. The polarization of pulse 1 and pulse 2 were copolarized, and that of pulse 3 was changed: parallel and cross polarizations to pulse 1 and pulse 2. The transient grating signal diffracted in the direction of $k_4 = k_1 - k_2 + k_3$ was recorded as a function of the time delay between pulse 2 and pulse 3 ($\Delta T_{23} = t$) when the time delay between pulse 1 and pulse 2 is 0 fs ($\Delta T_{12} = 0$). In order to measure the spectra of the diffracted light in the direction of k_4 at various time delays, we used a monochromator with the resolution of 0.06 nm connected to a charge coupled device camera. We also performed the photoluminescence (PL) measurement for the estimation of the exciton energies. The Ti:sapphire laser of the continuous wave mode with the wavelength of 800 nm was used for the excitation light, the detection system mentioned above also was used.

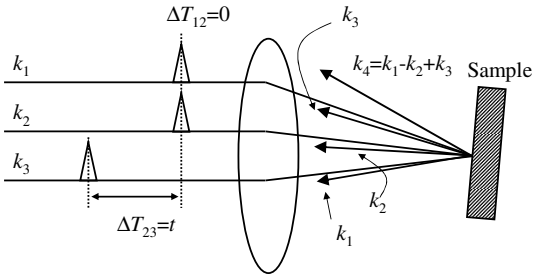

Fig. 1 Experimental configuration for a transient grating experiment. The pulse 3 is diffracted in the direction of k_4.

3 Results and discussion Figure 2 shows the transient grating signals of the sample.

Under the condition that pulse 3 is the parallel polarization to pulse 1 and pulse 2, the ultrafast response within 1.0 ps appears, while the time-domain signal shows only the single exponential decay in the case of the cross polarization. The full-width half maximum of the ultrafast optical response in the linear scale is about 250 fs, this ultrafast response has not been reported by the previous picosecond experiments. Both the transient grating signals have a rise time much longer than the pulse width and a decay time over 30 ps. The rise and decay times in the both signals estimated by fitting with a single exponential function are 4 and 120 ps, respectively. This decay time is shorter than the values reported in the multiple quantum well samples by the time-resolved photoluminescence-measurement [6].

In order to clarify the origins of the nonlinear optical response, the spectra of the transient grating signals were measured at various time delays. Figure 3 depicts the spectra of the transient grating signal observed at various time delays under the parallel-polarization condition in Fig. 2.

The top spectrum shows the PL spectrum. All spectra are normalized at the maximum intensity. The dashed and dotted lines indicate the energies of the heavy-hole (HH) and light-hole (LH) exciton states calculated by the model of the confinement of the center-of-mass motion. In the calculation, the homogeneous dead layer thickness of 95.6 nm is used as the thickness of the thin films [7]. The values of the effective masses in the calculation are taken from Ref. [8].

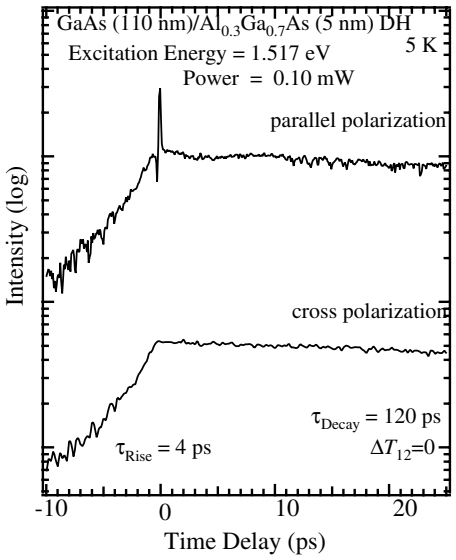

Fig. 2 Transient grating signals as a function of time delay ΔT_{23} at 5 K under $\Delta T_{12} = 0$. The upper and lower traces are detected under the condition that the polarization of the pulse 3 is parallel and cross to that of the pulse 1 and pulse 2, respectively.

All of the transient grating spectra obviously have several peaks around 1.516 eV, and are similar in shape to the PL spectrum. These peak energies agree with the PL-peak energies of the quantized excitonic states of weak confinement. This fact exhibits that the transient grating signals in Fig. 2 result from the nonlinear optical response of the excitons weakly confined in the GaAs thin film. The spectra have the

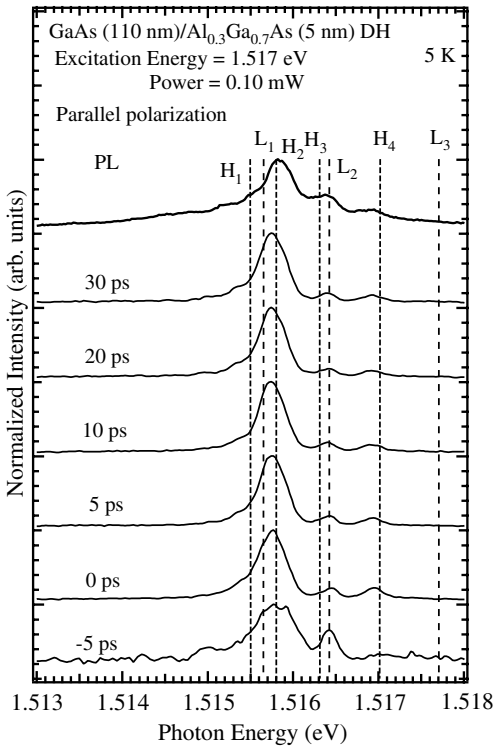

Fig. 3 Spectrum of the transient grating signals under the parallel polarization condition in Fig. 2 observed at various time delays. The top spectrum is PL spectrum. All spectra are normalized at the maximum intensity. The dashed and dotted lines indicate the energies of the quantized HH and LH exciton states due to the confinement of the center-of-mass motion. The notation of H_n (L_n) indicates the n-th quantized HH (LH) exciton state.

maximum intensity at the energy of the $n = 2$ HH exciton in all time delays, which is consistent with results by the picosecond-laser-pulse experiments that demonstrated the enhancement of the nonlinear optical response at the $n = 2$ HH-exciton energy [3–5]. In addition, the spectra show the peaks at the $n = 2$ LH-exciton and the $n = 4$ HH-exciton energies. The enhancement of the nonlinear optical response at these exciton energies has not been clearly observed in the picosecond-laser-pulse experiments.

Finally, we discuss the origin of the long decay time in the time-domain signals, phenomenologically. The decay time of the time-domain signals of 120 ps mentioned above is much longer than that evaluated from the radiative width of the $n = 2$ HH exciton. If the $n = 2$ HH exciton with the maximum intensity in the spectra only contributes to the nonlinear optical response, the transient grating signal should decay within about 10 ps corresponding to the spectral width of 0.4 meV. The femtosecond-laser pulse with the spectral width of 20 meV excites the free electrons and holes in addition to the excitons. Then, the free electrons and holes form not only the $n = 2$ HH exciton but also the higher excitons. Since the coupling of the higher exciton states with the radiation field is weak, the decay time of the higher exciton states are longer than that of the $n = 2$ HH-exciton state. In consequence, the nonlinear optical response under the femtosecond-laser-pulse-excitation condition may indicate the long decay time.

4 Conclusions We have demonstrated the nonlinear optical response of the excitons weakly confined in the GaAs thin films by a transient grating method under 160 fs pulse-excitation condition which has the spectral width of 20 meV. We find that the transient grating signals shows a long decay time of 120 ps and a rise time of 4 ps, and that the signal measured by the copolarized three pulses shows an ultrafast nonlinear optical response comparable to the pulse width. The spectra of the transient grating signals clearly show the peaks at the multiple exciton energies and maximum at the $n = 2$ HH exciton energy of the center-of-mass motion. This fact conclusively indicates that the characteristics of the nonlinear optical response of weakly confined excitons depend on the spectral width of the excitation-laser pulses.

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