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Ultrafast nonlinear optical response of weakly confined excitons in GaAs thin films

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We report on an ultrafast nonlinear optical response of weakly confined excitons in GaAs thin films observed by a two-beam degenerate four-wave-mixing (DFWM) technique. An ultrafast optical response comparable to the pulse width appears under ultrashort-pulse-laser excitation condition. We clarified from the DFWM spectra that the plural exciton states contribute to the ultrafast response. The results imply a possibility of ultrafast switching devices using weakly confined excitons.

1 Introduction The optical nonlinearity of excitons in semiconductors has been extensively studied towards realization of ultrafast optical devices including all-optical switching devices [1, 2]. Recently, an enhancement of the optical nonlinearity of weakly confined excitons in GaAs thin films due to the effect of nonlocality-induced-double resonance in energy and size (NIDORES) has been reported under the resonant excitation condition of a single exciton state by picosecond laser pulses [3,4]. The nonlocal response arises from the interaction of excitons with light fields in the case of the sample thickness close to the wavelength of light, and induces the fast population decay of excitons [5]. If the excitons under the NIDORES conditions demonstrate the ultrafast response, e.g., within 1 ps, ultrafast optical switching devices operating in sub-picosecond time scale may be realized. In the present work, we show ultrafast nonlinear optical responses of the weakly confined excitons in the GaAs thin films observed by a degenerate four-wave-mixing (DFWM) technique with various spectral widths of excitation pulses. By using an ultrashort pulse laser for the excitation, an ultrafast response comparable to the pulse width appears in the DFWM signals. The origin of the nonlinear optical response is discussed from the spectrally resolved DFWM signals.

2 Experiment The sample used in the present work is a double heterostructure (DH) thin film 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 films [3, 4]. The nonlinear optical response of the excitons was measured by a DFWM technique at 5 K. The experimental configuration is schematically shown in Fig.1. The light source was a mode-locked Ti:sapphire pulse laser with 76 MHz repetition. The center energy of the laser pulse was 1.5158 eV which agrees with the exciton energy in the present sample. The pulse train was split into two parts that were focused to a single spot of 100 μm in diameter on the sample. The polarization of the pulse 1 and pulse 2 were copolarized. The DFWM signals in the backward direction of $2k_2 - k_1$ were measured by various

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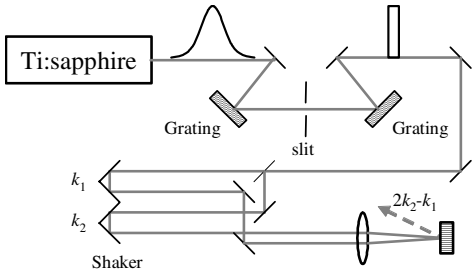

Fig. 1 Experimental configuration for a DFWM method. The spectral width of the excitation pulse was controlled by a slit between a grating pair.

spectral widths of the excitation pulse, ranging from 0.8 to 19.2 meV without changing the center energy. The spectral width of the laser was controlled by a slit between the grating pair. The excitation density was kept at 12 nJ/cm^2 in all the spectral width. To measure the DFWM spectra at various time delays, we used a monochromator with a 0.06-nm resolution connected to a charge-coupled device camera. We also performed photoluminescence (PL) measurements for the estimation of the exciton energies. The Ti:sapphire laser of the continuous wave mode of 800 nm was used for the excitation light, the detection system mentioned above was also used.

3 Results and discussion Figure 2 shows the DFWM signals observed by various spectral widths of the laser pulses. Each DFWM signal is normalized at the maximum intensity. At the spectral width of 0.8 meV, the signal-decay time estimated from fitting by a single exponential function is 1.6 ps which is induced by the NIDORES effect. The asymmetric profile results from the temporal distortion of the excitation pulse owing to the spectral control system. In order to remove this temporal distortion around 0 ps, the estimation of decay time was performed for the signal in the time region after 5 ps. By excitation using spectrally broader laser pulses, the ultrafast response comparable to the pulse width appears around 0 ps in addition to the decay component with the time constant of 1.3 ps which almost agrees with the decay time of the weakly confined excitons due to the NIDORES effect.

In order to clarify the origins of the ultrafast nonlinear optical response, the DFWM spectra under the excitation of the spectral width of 19.2 meV have been measured at various time delays. The DFWM spectra at various time delays are depicted in Fig. 3(b). In addition, the PL spectrum is shown in Fig. 3(a) for reference, where the dotted lines indicate the exciton energies. The origins of the peaks were assigned by comparison with the calculation results based on the quantization of the center-of-mass motion of excitons. The notation of HHn (LHn) means the n-th quantized heavy- (light-) hole exciton state. The optical transition is described by the nonlocal response regime instead of the long-wavelength approximation when the film thickness is close to the wavelength of the light. In the nonlocal response regime, the phase difference between the light wave and exciton wave function should be considered, and the transition probability is determined by the integral of exciton wave functions multiplied by the resonant electromagnetic field of light. Therefore, in the case of the film thickness of 110 nm, the only excitons with even quantum number have the peaks [3, 4].

The DFWM spectra clearly show several peaks at the exciton energies around 0 ps (thick curve) in addition to the major peak of the HH2-exciton energy. The intensity of those excitonic peaks decreases within a few hundred femtoseconds, which corresponds to the ultrafast signal decay in Fig. 2. These results indicate that the ultrafast response is related to the response of the weakly confined excitons. It is well known that the DFWM-signal decay reflects the dephasing time. As possible factors for causing the ultrafast response, the population decay and the pure dephasing of the excitons should be considered. The fact that we observed the long decay time over 100 ps under the femtosecond pulse excitation in our previous report [6] indicates negligible contribution of the population decay to the DFWM-signal decay. Therefore, we focused only on the exciton dephasing. On the other hand, the DFWM spectra around 0 ps have sharp peaks with a full width half maximum of 0.4 meV, which denies the possibility of ultrafast

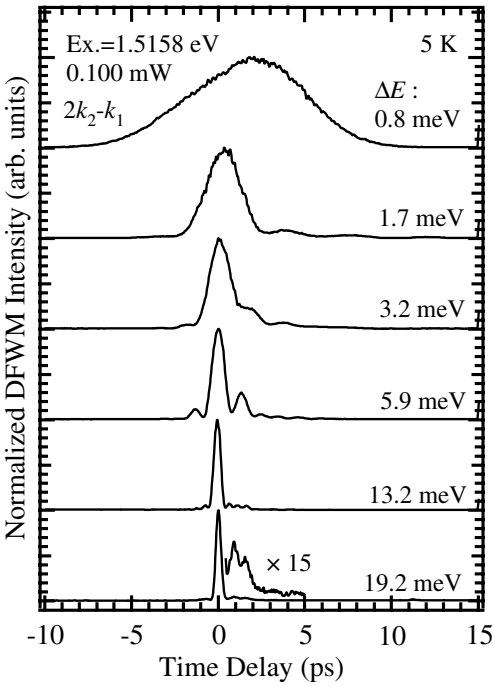

Fig. 2 DFWM signals observed by various spectral widths of the laser pulses. All signals are normalized at the maximum intensity.

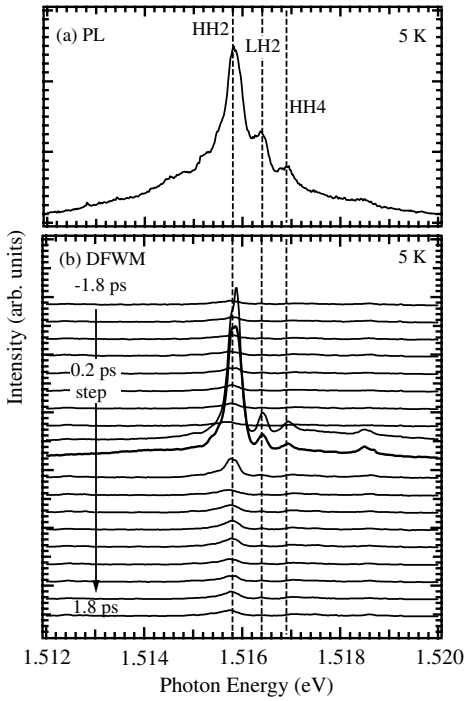

Fig. 3 (a)PL spectrum. (b)DFWM spectra observed at various time delays by using the laser pulse with spectral width of 19.8 meV. The dotted lines indicate the energies of the quantized excitons. The thick curve is the spectrum at 0 ps.

exciton dephasing. As mentioned above, we observed a dephasing time of 1.3 ps around tail of the DFWM signal which is attributed to the exciton dephasing. The exact origin of this ultrafast decay in the GaAs thin

films is still unclear. Zhang et al. reported a possibility of the contribution of exciton-exciton scattering to the ultrafast decay process in the ZnO thin films [7]. Even in the low excitation density the ultrafast response of weakly confined excitons was observed (not shown here), the scattering process may not be a major factor for the response. Moreover, the off-resonant excitation of continuum states may induce the destructive interference of excitons, leading to the rapid decay. However, the DFWM signal at 1.7 meV, which is less than the exciton binding energy, shows a decay comparable to the pulse width in Fig. 2. Hence, the ultrafast response is almost independently of the excitation of the continuum state. One of the possible reasons is interference between the diffracted light waves. Namely, the interference between the waves from each exciton state under the excitation condition of plural exciton states may lead to the ultrafast nonlinear optical response.

4 Conclusions We have investigated the nonlinear optical response of weakly confined excitons in GaAs thin films by the DFWM method with various spectral widths under the NIDORES condition. Under the irradiation of ultrashort laser pulses, we have found that a ultrafast nonlinear optical response comparable to the pulse width appears and that the light waves from the plural exciton states contribute to the nonlinear optical response in the DFWM spectra. From these results, we expect that the NIDORES effect on weakly confined excitons leads to a novel ultrafast optical switching device.

References

[1] F. T. Vasko and A. V. Kuznetsov, Electronic States and Optical Transitions in Semiconductor Heterostructures: Graduate Texts in Contemporary Physics (Springer-Verlag, New York, 1999).
[2] J. Shah, Ultrafast Spectroscopy of Semiconductors and Semiconductor Nanostructures, 2nd ed., edited by M. Cardona, Springer Series in Solid-State Sciences, Vol. 115 (Springer-Verlag, Berlin, 1999).
[3] K. Akiyama, N. Tomita, Y. Nomura, and T. Isu, Appl. Phys. Lett. **75**, 475 (1999).
[4] H. Ishihara, K. Cho, K. Akiyama, N. Tomita, Y. Nomura, and T. Isu, Phys. Rev. Lett. **89**, 17402 (2002).
[5] K. Akiyama, N. Tomita, T. Nishimura, Y. Nomura, T. Isu, H. Ishihara, and K. Cho, Physica B **314**, 293 (2002).
[6] O. Kojima, T. Isu, J. Ishi-Hayase, and M. Tsuchiya, phys. stat. sol. (c) **3**, 675 (2006).
[7] W. Zhang, H. Wang, K. S. Wong, Z. K. Tang, G .K. L. Wong, and R. Jain, Appl. Phys. Lett. **75**, 3321 (1999).

