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Spectral width dependence of residual carrier effect on nonlinear optical response of weakly confined excitons

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We report residual carrier effects on the nonlinear optical response of weakly confined excitons in GaAs thin films. The nonlinear response was investigated using a degenerate four-wave mixing technique with an additional pulse to create the residual carriers. In the case of the additional-pulse incidence, the signal intensity decreases with the time of incidence. However, the ultrafast component comparable to the pulse width in the temporal profiles does not change appreciably. Our results imply the possibility of the realization of ultrafast switching using the exciton response.

1 Introduction Ultrafast optical switches based on optical nonlinearity of excitons in nanostructured semiconductors will be one of the key devices for the development of future ultrafast all-optical networks. Recently, there have been reports on the enhanced optical nonlinearity of weakly confined excitons resulting from coupling with light fields under the condition of nonlocality-induced double resonance in energy and size (NIDORES) in 110 nm thick GaAs thin films [1, 2]. In the weak confinement condition, in which a confinement space is larger than an exciton Bohr radius and is of the same order as light wavelength, the exciton is characterized by the nonlocal response theory [3, 4]. In a previous report, using a degenerate four-wave mixing (DFWM) tecquniqe, we have shown that the ultrafast response of excitons under the NIDORES condition originates from the interference effect between exciton states [5]; this indicates a potential for ultrafast response devices. On the other hand, because the enhancement of the optical nonlinearity due to the NIDORES effect requires a high sample quality to induce the exciton-light coupling, the excitons have long lifetime [6]. This is disadvantageous for the realization of ultrafast response devices with a high repetition rate such as 1 Tbit/s, because residual carriers may disturb the exciton response. In the present work, we study the residual carrier effect generated by an optical pulse on the exciton response under the NIDORES condition in GaAs thin films. We observed DFWM signals for the pulses of various excitation spectral widths with an additional pulse to create residual carriers. Although the DFWM signal intensity decreases with the additional pulse, the response time is not affected appreciably by the additional pulse. Our results indicate the potential for ultrafast optical switching by using weakly confined excitons. We discuss the effect of the residual carriers upon the nonlinear optical response.

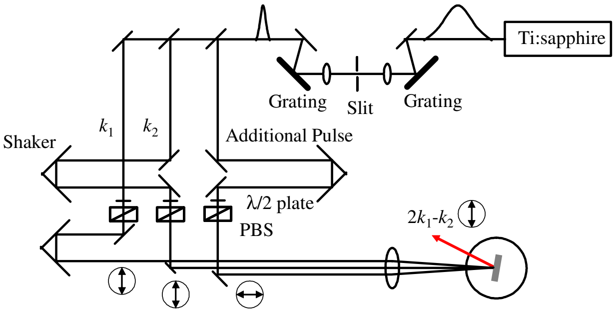

Figure 1 Schematic experimental setup. The arrows in circles indicate the polarization direction of each pulse.

2 Experimental The sample used in the present work is a double heterostructure (DH) with three periods of GaAs (110 nm)/$Al_{0.3}Ga_{0.7}As$(5 nm) on a (001) 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 [5,7]. The nonlinear optical response was measured using a DFWM technique at 3.5 K in the backward direction of $2k_1 - k_2$. Figure 1 shows the schematic of the experimental setup. A mode-locked Ti:sapphire pulse laser with a pulse width of 160 fs was used as the light source. The laser photon energy was tuned to 1.5158 eV which is the confinement quantum number $n = 2$ confined exciton energy with maximum optical nonlinearity. The spectral width of the excitation pulse ($\varDelta E$) was changed using a slit between the gratings. To create residual carriers, an additional pulse enters the sample at various delay times in different incident direction from k_1 and k_2. Moreover, in order to eliminate the overlap of the DFWM signal induced by the additional pulse in the $2k_1 - k_2$ direction, the polarization of additional pulse was orthogonal to those of pulse 1 and pulse 2, as shown in Fig. 1. Hence, the DFWM signal arise only around zero delay. The light power of each pulse was kept at 1.2 pJ/pulse for 100 μm diameter spot size corresponding with an excitation density of 12 nJ/cm^2. Below this excitation power, the DFWM intensity shows a cubic dependence on the laser power [1].

3 Results and discussion Figure 2 shows the effect of the additional pulse on the DFWM signal of the weakly confined excitons at $\varDelta E$=16 meV. The DFWM signals originate from the optical nonlinearity of the excitons confined in the thin films [5, 7]. The dotted curve is the standard signal without the additional pulse. The time of incidence of the additional pulse (e.g., 50 ps) with respect to time zero is indicated using arrows. The negative (positive) time corresponds to the additional-pulse incidence before (after) the generation of DFWM signals. All signals indicate an ultrafast response comparable to the pulse width arising from the interference effect of exciton states. The decay component near the signal tail arises from exciton dephasing. The maximum DFWM intensity indicated by the arrows varies with the time of incidence of the additional pulse; however, the temporal waveform, i.e., the response time, does not change appreciably with the time of incidence. This is an advantage for ultrafast switching applications.

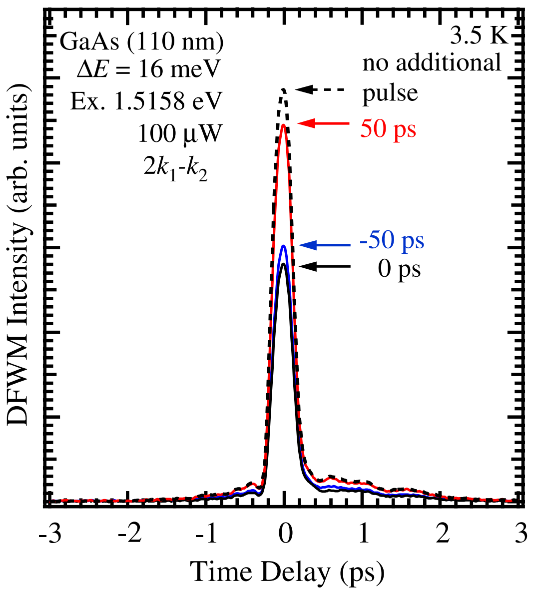

Figure 2 DFWM signals at $\varDelta E = 16$ meV under various additional pulse incident condition. The arrows indicate the maximum intensity of each signal. The times next to the arrows indicate the times of incidence.

The signal intensity decreases to 62% when an additional pulse is incident at −50 ps, and to 58% at zero delay. Moreover, although the signal intensity in the case of +50 ps time delay recovers, it recovers only 90%. The variation of time of incidence corresponds to that of the residual exciton density. Under the condition of ultrashort-pulse irradiation, the weakly confined excitons have a lifetime component of over 200 ps [5]. Therefore, the excitons created at the negative time region are present at zero delay, and lead to a decrease in the DFWM intensity by 60%. In addition, the decrease in the signal intensity in the case of the additional pulse incident in the positive time region suggests the existence of a long-lifetime component over a few nanoseconds. In fact, we reported a long decay time of 14 ns using time-resolved photoluminescence under non-resonant excitation conditions. Thus, the excitons created at the positive time region are present after 13 ns ($\sim$ 76 MHz), and reduce the signal intensity [6].

Figure 3 demonstrates DFWM signal dependence on $\varDelta E$ of the additional pulse incidence. For each $\varDelta E$, all signals are normalized using the standard signal without the additional pulse indicated by the dotted curve. An exciton response comparable to the pulse width and the oscillatory structure due to interference between exciton states appear.

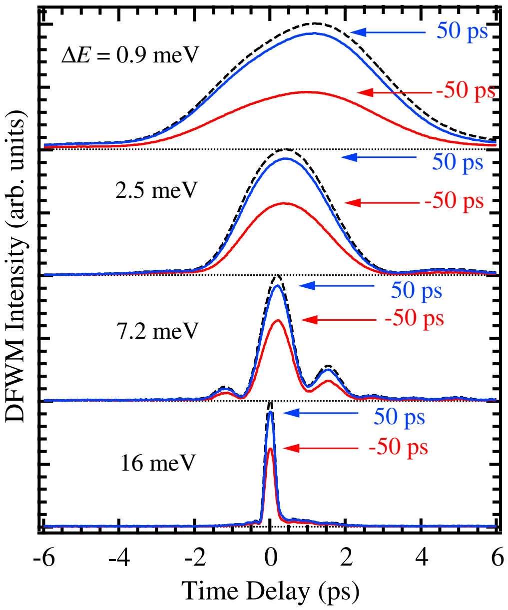

Figure 3 DFWM signal dependence on ΔE of additional pulse incidence. The dotted curves are standard signals for each ΔE.

All profiles demonstrate the same characteristics: although the intensity decreases to approximately 60% in the negative time region and to 90% in the positive time region, the temporal profiles have almost the same shape.

In Fig. 4, the intensity ratio of the DFWM signal is plotted as a function of the time of incidence of the additional pulse with varying ΔE. Although the reduction rate in the positive time region is almost constant, that in the negative time region clearly depends on ΔE. In case of $\Delta E = 0.9$ meV, the signal intensity is lower than 50% of that without the additional pulse. This ΔE dependence of signal intensity in the negative time region results from the creation efficiency of the excitons. As ΔE is closer the exciton line width, excitons are created more efficiently and the residual carrier density increases.

4 Conclusion

We have investigated the residual exciton effects on the nonlinear optical response of weakly confined excitons under various ΔE conditions. Using the additional-pulse incidence to create residual excitons, the DFWM signal intensity decreased with the variation in residual exciton density. However, the temporal shape of an ultrafast response comparable to the pulse width did not change significantly with the incidence, which is an advantage for ultrafast operating devices. We concluded that our results indicate the possibility of ultrafast switching using excitons under the NIDORES condition.

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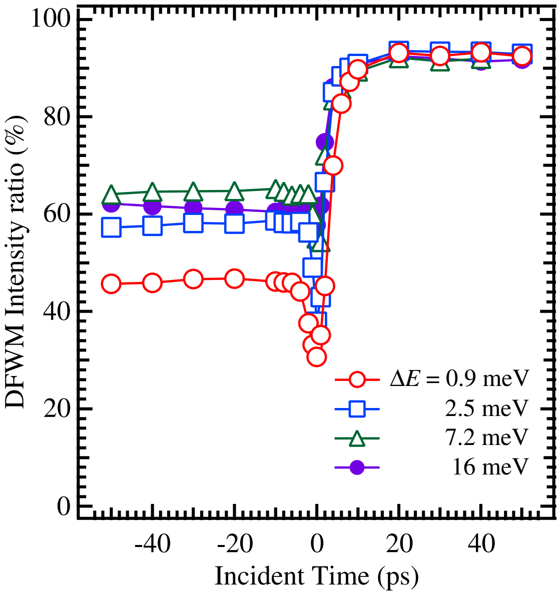

Figure 4 Intensity ratio of the DFWM signal under various ΔE conditions.

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