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# Effects of excitation spectral width on decay profile of weakly confined excitons

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**Abstract**

We report the effect due to a simultaneous excitation of several exciton states on the radiative decay profiles on the basis of the nonlocal response of weakly confined excitons in GaAs thin films. In the case of excitation of single exciton state, the transient grating signal has two decay components. The fast decay component comes from nonlocal response, and the long-lived component is attributed to free exciton decay. With an increase of excitation spectral width, the nonlocal component becomes small in comparison with the long-lived component, and disappears under irradiation of a femtosecond-pulse laser with broader spectral width. The transient grating spectra clearly indicates the contribution of the weakly confined excitons to the signal, and the exciton line width hardly changes by excitation spectral width. From these results, we concluded that the change of decay profile is attributed not to the many-body effect but to the effect of simultaneous excitation of several exciton states.


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

## 1. Introduction

Nonlinear optical response of excitons in nanostructured semiconductors, e.g. quantum wells and quantum dots, has been interesting for application of optical devices. For the realization of such devices using the exciton response, the optical nonlinearity and the exciton lifetime are important factors. Recently, the control of those factors resulting from the nonlocality-induced double resonance in energy and size (NIDORES) effect has been reported by using a picosecond pulse laser [1–3]. Under the NIDORES condition, excitons are weakly confined in the GaAs thin films with a thickness much larger than the exciton Bohr radius and characterized by the nonlocal response theory [4,5]. For the realization of ultrafast-optical-response devices by NIDORES effects, the usage of the ultrashort-pulse laser with broad spectral width is needed. Since broadband excitation leads to a simultaneous excitation of several exciton states, it may induce a response different from the response by a single exciton state.

In the present work, we have investigated the relation between the excitation spectral width and the exciton decay profile under the NIDORES condition by a transient grating (TG) technique. We have found that the profile of the TG signal is changed by the spectral width of the excitation pulse. Especially, the decay component due to the nonlocal response disappears under irradiation of the ultrashort pulse with broader spectral width. However, in the TG spectra, the exciton profiles in the energy domain hardly change by the excitation spectral width. We discuss the change of the decay profile from the viewpoint of the excitation effect of several exciton states.

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## 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 [1–3]. In the case of film thickness of about 110 nm, an enhancement of the nonlinear optical response of the $n=2$ confined exciton due to the NIDORES effect has been reported [1,2]. The exciton-population decay was measured by a TG technique at 5 K in the time and spectral regions in the backward direction of $k_1-k_2+k_3$. The experimental configuration is schematically shown in Fig. 1. The laser source was a mode-locked Ti:sapphire pulse laser delivering 170-fs pulse with a repetition rate of 76 MHz. By a slit between a grating pair, the laser spectral width $\Delta E$ was controlled in the range 0.9–15.3 meV without changing the center energy of 1.5158 eV, which is the resonant energy of the $n=2$ confined exciton showing maximum optical nonlinearity in the sample. The excitation power was kept at 100 μW, corresponding to the excitation density of 12 nJ/cm$^2$. The intensity ratio of the power of excitation ($k_1$ and $k_2$) and probe pulses ($k_3$) is 10:1. The polarization of $k_1$ and $k_2$ were copolarized, and that of $k_3$ was cross-polarization to the two excitation pulses. In the TG measurement, 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. To measure the TG spectra, we used a monochromator with a 0.06-nm resolution connected to a charge-coupled device camera.

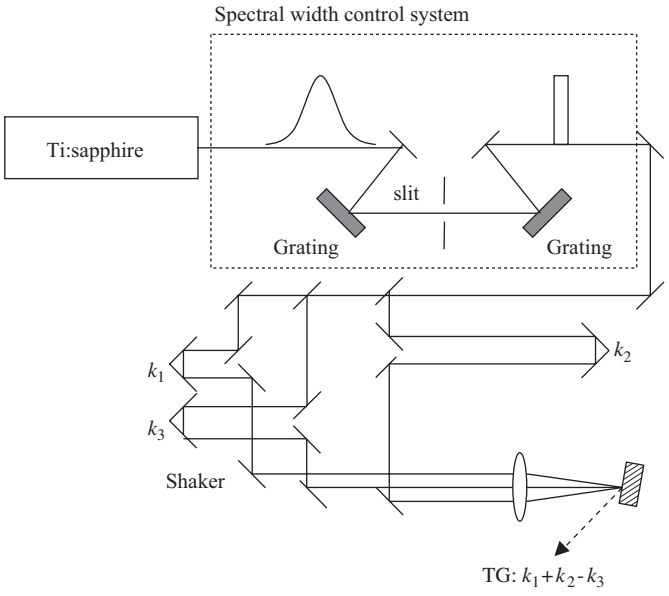


Fig. 1. Experimental configuration for a TG method. The spectral width of the excitation pulse was controlled by a slit between a grating pair.

## 3. Results and discussion

Fig. 2 shows the $\Delta E$ dependence of TG signal. Initially, we focus on the TG signal at $\Delta E = 0.9$ meV. In the case of $\Delta E = 0.9$ meV, it is regarded that the laser pulse almost excites a single weakly confined exciton state. The TG signal has two decay components. The fast component is due to the radiative decay process arising from the coherent processes based on the nonlocal response [2], and the long-lived one is attributed to a free exciton relaxation [6].

With an increase of $\Delta E$, the fast component becomes small and disappears at $\Delta E = 15.3$ meV, corresponding to excitation by femtosecond pulse laser. In a previous work, we have reported that the weakly confined exciton excited by the ultrashort pulse laser contributed to TG signal in all the time regions [7]. Hence, the disappearance of the fast component may be attributed to the loss of coherent radiative process by the nonlocal response.

In order to reveal the origins of the change of the decay profile, we measured the TG spectra at various $\Delta E$. Fig. 3 indicates the TG spectra at zero delay under various $\Delta E$. The laser spectra are depicted by dotted curves. Every TG spectra show a sharp peak at the same energy of 1.5158 eV, which is the energy of the $n=2$ weakly confined exciton. The $n=2$ confined exciton is the lowest state which is optically allowed according to the nonlocal response in our sample [3,7]. The full-width at half-maximum of the TG

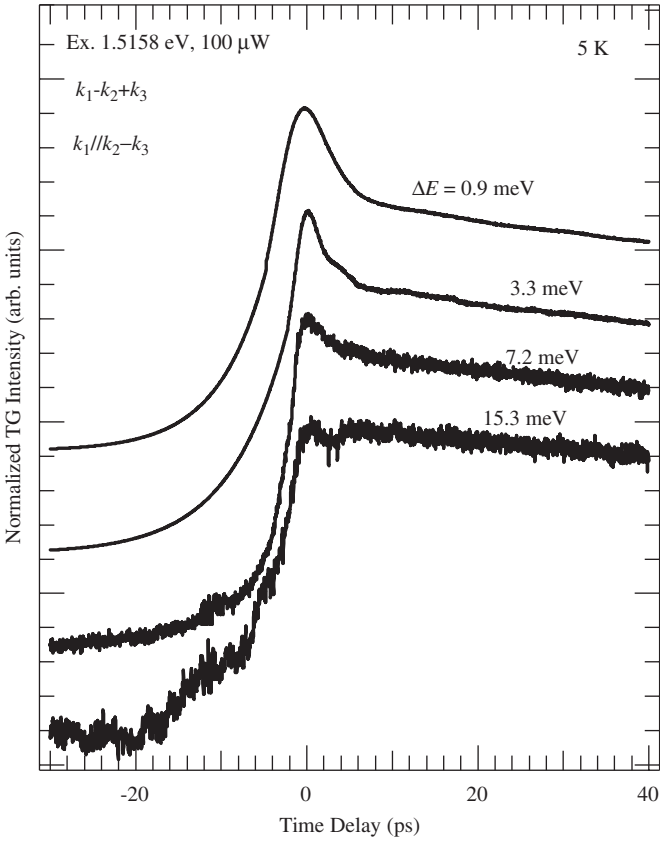


Fig. 2. TG signals observed at various $\Delta E$. All signals are normalized at the maximum intensity.

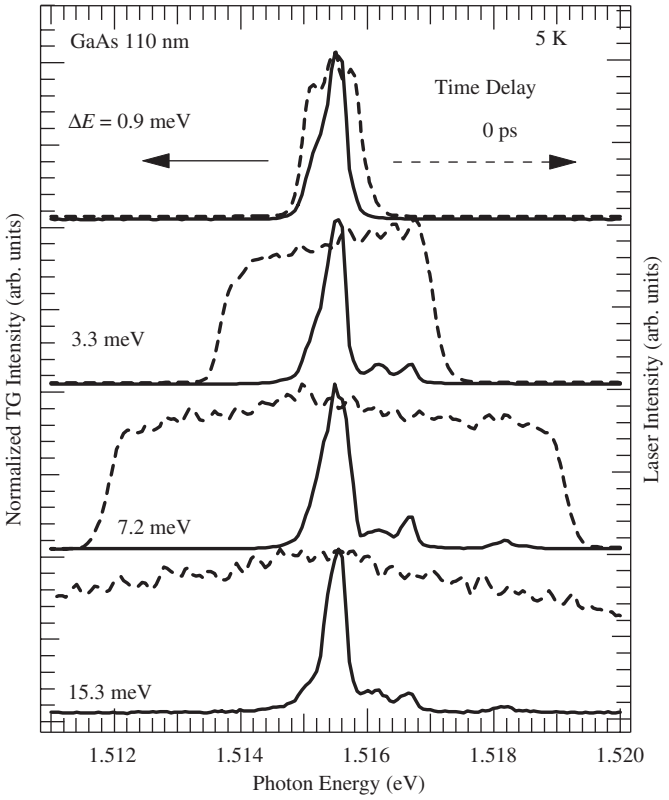


Fig. 3. $\Delta E$ dependence of TG spectrum (solid curves) observed at zero delay. The dotted curves are laser profiles.

spectra is almost the same width for every $\Delta E$ even at $\Delta E \approx 15.3$ meV. The disappearance of the fast component in the time-domain signal cannot be explained by the high-density excitation effects. If an increase of exciton density results in the exciton screening and the exciton–exciton scattering effects, the TG spectrum will show an exciton energy shift and broadening of the exciton line width. However, the results showed the same spectral shape. Thus, we concluded that multi-exciton effects due to an increase of excitation density with decreasing pulse width hardly contribute to the disappearance.

In the case of $\Delta E \geqslant 3.3$ meV, the TG spectra display higher order exciton peaks, which indicates simultaneous excitation of several exciton states. This simultaneous excitation induces an interaction between the weakly confined exciton states. Indeed, we observed the interference between the exciton states as quantum beats in degenerate four-wave-mixing signals [8]. Therefore, our results demonstrate the interaction between the exciton states due to the simultaneous excitation of several exciton states. The effect results in the fast vanishing of the coherent processes and changes in the exciton decay profiles.

## 4. Conclusion

We have investigated the exciton relaxation by a TG technique with varying spectral width. The exciton decay profile shows two components due to nonlocal coherent processes and the free exciton decay under an excitation of single exciton state. The decay component due to the nonlocal response decreases with increasing excitation spectral width. From the TG spectra under various $\Delta E$, we deduced that the change of the exciton relaxation profile is caused by an interaction between several exciton states excited by an ultrashort pulse with broad spectral width.

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