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Enhancement of nonlinear optical response of weakly confined excitons in GaAs thin films by spectrally rectangle-shape-pulse-excitation

O Kojima^{1,2,3}, T Isu^{1,2,4}, J Ishi-Hayase^{1,2}, M Sasaki¹ and M. Tsuchiya^{1,2}

¹ National Institute of Information and Communications Technology, 4-2-1, Nukui-Kitamachi, Koganei, Tokyo, 184-8795 Japan

² CREST, Japan Science and Technology Agency

E-mail: kojima@phoenix.kobe-u.ac.jp

Abstract. We report the enhancement of the nonlinear optical response of the weakly confined excitons with use of spectrally rectangular pulse. The nonlinear optical response was investigated as a function of excitation energy by a degenerate four-wave-mixing (DFWM) technique. In the case that the laser pulse with the controlled spectral shape excites the plural exciton states simultaneously, the DFWM signal intensity is enhanced by a factor of two in comparison with the intensity under the excitation of a single exciton state. This enhancement is caused by the superposition of the nonlinear optical responses from the plural exciton states.

1. Introduction

Nonlinear optical response of excitons in semiconductors has been extensively studied from the aspects of the exciton dynamics and application to ultrafast optical devices [1, 2]. Recently, enhancement of the optical nonlinearity of the weakly confined excitons in semiconductor thin films due to strong coupling with radiation fields has been reported in the degenerate four-wave-mixing (DFWM) measurement, which is called the nonlocality-induced-double resonance in energy and size (NIDORES) [3, 4]. In the weakly confinement regime, the center-of-mass motion of the exciton is quantized, and the coherent extension of the wavefunction of each state is coupled with the radiation field. By the nonlocal theory, the large nonlinear response of the spatially extended "nondipole-type" excitonic state is caused by the size-resonant enhancement of the internal field with a nanoscale spatial structure. These experimental results were based on a selective excitation of a single exciton state with use of a pulse laser with the temporal width of the order of picosecond. However, if a femtosecond-pulse laser with the broader spectral width is used to obtain the faster response of excitons, the detuning components of the laser spectrum for the exciton resonance energy will be larger. Then, the DFWM intensity will decrease with increasing off-resonance components [5], which is a problem for realization of

³ Present address: Department of Electrical and Electronics Engineering, Kobe University, 1-1, Rokkodai, Nada, Kobe 657-8501, Japan

⁴ Present address: Center for Frontier Research of Engineering, The University of Tokushima, 2-1 Minamijosanjima-cho, Tokushima 770-8506, Japan

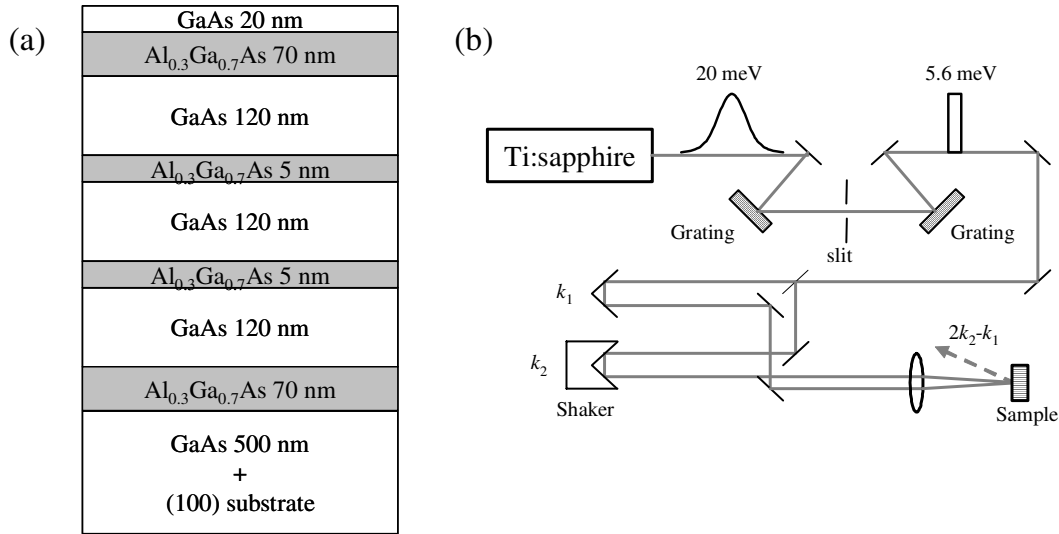


Figure 1. (a)Sample structure. (b)Schematic configuration of a DFWM experiment. The sample structure is controlled by the grating pair and a slit

the optical device showing an ultrafast and intense nonlinear optical response. If the spectral shape of the excitation pulse is adequately controlled and the plural exciton states under the NIDORES condition are simultaneously excited, one can expect more intense DFWM signal due to the superposition of the nonlinear optical responses of the optically generated excitons. In the present work, we propose the further enhancement method of the nonlinear optical response of weakly confined excitons under the plural exciton-excitation condition by using a DFWM technique of laser pulses with spectrally-rectangular shape. It has been found that the DFWM intensity is enhanced twice larger than that under the excitation of a single exciton state in the case that the laser pulse with the controlled spectral shape excites the plural exciton states.

2. Experiment

The sample used in the present work is the double heterostructure thin films with 3 periods of GaAs(120 nm)/Al_{0.3}Ga_{0.7}As(5 nm) on a (100) GaAs substrate grown by molecular beam epitaxy, as shown in Fig. 1(a). 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 the thickness around 110 nm, the enhancement of the nonlinear optical response has been reported [3, 4]. The nonlinear optical response of excitons was measured by a DFWM method at 5 K. The light source was a mode-locked Ti:sapphire pulse laser with repetition of 76 MHz. The spectrally rectangular shape of the laser pulse was extracted from the spectrally broader femtosecond-laser-pulse by a slit located at the center of the grating pair, as schematically shown in Fig. 1 (b). The laser energy was changed in the exciton-energy region without changing the typically spectral width (full width half maximum) of 5.6 meV. The temporal width of extracted pulse was 525 fs. The pulse train is split into two parts that are focused to a single spot on the sample. The separated two beams were parallel polarized. The excitation power was kept at 100 μ W. The DFWM signals diffracted in the direction of $2k_2-k_1$ were recorded. Our experiments were performed with a fast-scan setup without using a chopper for improvement of the signal-to-noise ratio. Moreover, in order to estimate the energies of the weakly confined excitons, we measured the DFWM spectrum by a monochromator with the spectral resolution of 0.15 nm connected to a charge coupled device camera.

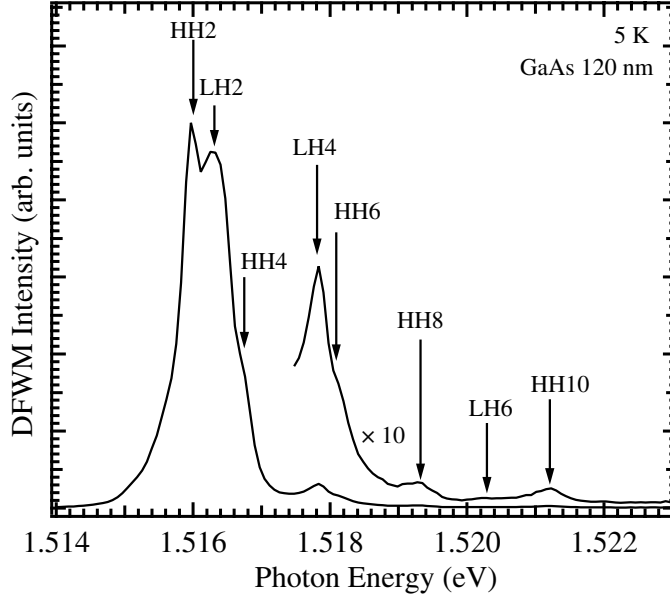


Figure 2. DFWM spectrum in the GaAs sample. The notations of HHn (LHn) indicate the n -th heavy-hole (light-hole) excitons.

3. Results and Discussion

Figure 2 shows the DFWM spectrum in the GaAs sample. In the measurements, the pulse with the spectral width of 10 meV was used to excite in the broader energy region. Many excitonic DFWM peaks are observed. The origins of the peaks were estimated from the calculation based on the model of the confinement of the exciton center-of-mass motion, where the notations of HHn (LHn) indicate the n -th heavy-hole (light-hole) excitons. The optical transition is described by the nonlocal 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 waves of the light and exciton should be considered, and the transition is determined by the integral of the wave function of excitons multiplied by the resonant electromagnetic field of light. Therefore, the only excitons with even quantum number have the peaks due to the nonlocal response in the case of the thickness of 120 nm [3, 4].

The DFWM signals measured at various excitation energies are depicted in Fig. 3(a), where the energies indicate the center energy of the laser pulse. The intensity of the DFWM signal clearly depends on the excitation energy. The oscillatory structure in the signals is attributed to the quantum beat of the weakly confined excitons. For clarification of the excitation-energy dependence of the DFWM intensity, we pay attention to the intensity at 0 ps in the present work. We plotted the peak intensity of the DFWM signal at 0 ps as a function of excitation energy in Fig. 3(b). The DFWM signal intensity increased with an increase in excitation energy below the energy of 1.5193 eV. This excitation-energy dependence was quite different from that using ordinary Gaussian pulses, which only the exciton-resonance peak was observed at the $HH2$ -exciton energy [3].

In order to clarify the increment mechanism of the nonlinear optical response, we compared the result of the DFWM measurement with the spectrum. In the case of the excitation energy of 1.5143 eV, the $HH2$, $LH2$, and $HH4$ excitons are excited by the spectrally rectangle pulse. At 1.5150 eV, the higher-energy-edge of the laser pulse achieves at the $LH4$ -exciton energy, and

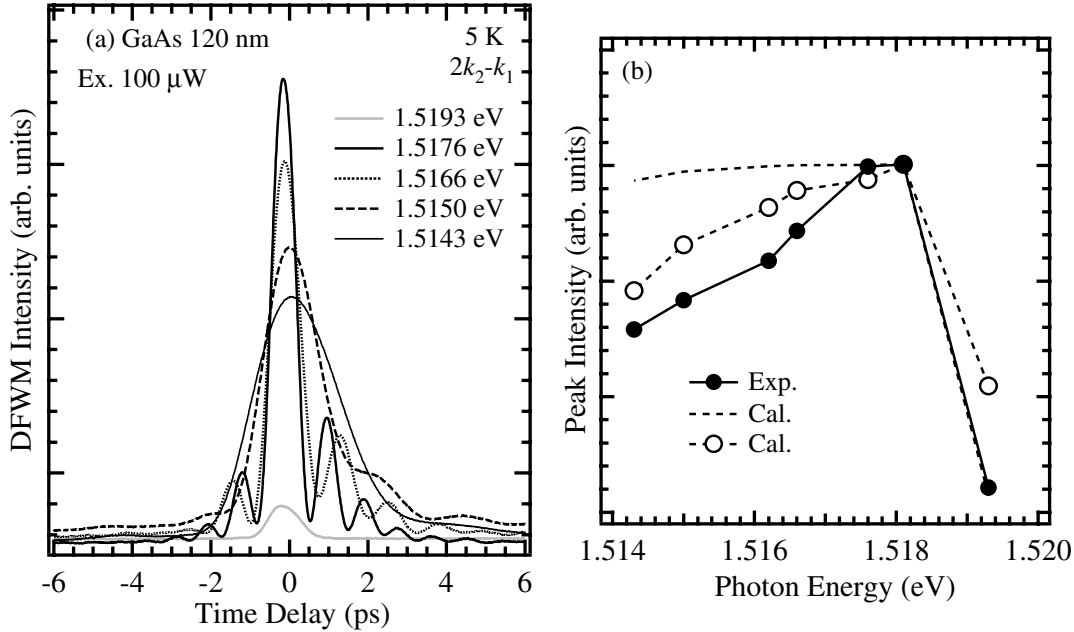


Figure 3. (a)DFWM signal in the GaAs sample. (b)The excitation energy dependence of the peak intensity of DFWM signal. The dotted curves are the estimated DFWM intensities.

then, the edge reaches to the HH10 exciton at the excitation energy of 1.5176 eV. Therefore, we assumed that the increment arises from the superposition of nonlinear optical response of the excited plural higher exciton states. Since the laser pulse at the excitation energy of 1.5193 eV detunes to the $n = 2$ exciton with maximum optical nonlinearity, the signal intensity rapidly decreases. In the case that the Gaussian pulse is used in the DFWM measurement, the DFWM intensity will rapidly decrease under the condition of detuning to the $n = 2$ exciton even if the same spectral width because the excitation intensity markedly decreases. Hence, the effect of superposition of the DFWM intensity of each exciton state is small. In our measurement, the rectangular pulse efficiently excites plural exciton states, and the spectral limitation of the laser pulse decreases the off-resonant component. Therefore, the nonlinear optical response is enhanced due to the superposition.

However, it is obvious that the superposition process is nonlinear. The conclusive process is unclear, and the experiments to investigate this process are underway. One of the possible reasons is the superposition of the diffracted waves. If the DFWM-signal intensity at 0 ps is given by the simply summation of the DFWM intensity of each exciton state, the increment slope in Fig. 3(b) should be smaller as shown by the dotted curve, where the DFWM intensity of each exciton state was estimated from the comparison the spectral intensity of each exciton state with that of the HH2 exciton in Fig. 2, and the intensity in the calculation is normalized by the maximum value of the experimental data. Then, we presumed that the DFWM intensity of each exciton state does not independently contribute to the intensity, and that the superposition of the diffracted waves by each exciton state leads to the intensity. Since the intensity is in proportion to the square of the amplitude of the light wave, we estimated the intensity: $I_{\text{DFWM}}(1.5143\text{eV}) \propto (\sqrt{I_{\text{HH2}}} + \sqrt{I_{\text{LH2}}})^2$, $I_{\text{DFWM}}(1.5150\text{eV}) \propto (\sqrt{I_{\text{HH2}}} + \sqrt{I_{\text{LH2}}} + \sqrt{I_{\text{HH4}}} + \sqrt{I_{\text{LH4}}})^2$, \dots , $I_{\text{DFWM}}(1.5176\text{eV}) \propto (\sqrt{I_{\text{HH2}}} + \sqrt{I_{\text{LH2}}} + \dots + \sqrt{I_{\text{HH10}}})^2$, where $I_{\text{DFWM}}(E)$ is the observed DFWM-signal intensity at the excitation energy of E , and $I_{\text{HH(LH)}}_n$ gives the DFWM

intensity at the HH(LH) n -exciton energy. The calculation result is demonstrated by the dotted curve with open circle in Fig. 3(b). The intensity in the calculation is normalized by the maximum value of the experimental data. The calculated profile modestly agrees with the experimental results. Therefore, the superposition of the diffracted waves may contribute to the DFWM-signal intensity. For more strict interpretation, we should consider the results by using the other parameters, e.g., oscillator strength. Our findings exhibit that the excitation by the spectrally-rectangle-shaped pulse has the great advantage for the enhancement due to the superposition of the optical nonlinearities of plural exciton states, namely, the usage of the laser pulse with adequate spectral width leads to the ultrafast response and the intense optical nonlinearity. Therefore, the rectangle-shaped pulse may give some insights for the realization of the ultrafast switching device.

4. Conclusion

We have investigated the nonlinear optical response of the weakly confined excitons in the GaAs thin films by a DFWM technique with use of spectrally rectangle pulse. We find that the DFWM intensity depends on the excitation laser energy, the maximum DFWM intensity is about twice larger than the case of the excitation of only the HH2 exciton which strongly contributes to the nonlinear optical response in the nonlocal response regime. In this enhancement process, the number of the exciton states included in the laser spectrum is the important factor. It is concluded from the result described above that the further enhancement in addition to the NIDORES effect arises from the superposition of the nonlinear optical response of plural exciton states. This gives some insights for the realization of the ultrafast switching devices without decrease of the DFWM intensity by using femtosecond pulse laser.

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