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Kojima, Osamu
Hayashii, Ken
Kita, Takashi
Akahane, Kouichi

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Pulse modulation towards low power operation based on quantum beat of excitons in a GaAs/AlAs multiple quantum well

Osamu Kojima[1], **Ken Hayashii**[1], **Takashi Kita**[1], **Kouichi Akahane**[2]

[1]Department of Electrical and Electronic Engineering, Graduate School of Engineering, Kobe University, 1-1 Rokkodai, Nada, Kobe 657-8501, Japan
[2]National Institute of Information and Communications Technology, 4-2-1 Nukui-kitamachi, Koganei, Tokyo 184-8795, Japan

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

Abstract. We report pulse modulation caused by the exciton quantum beat in a GaAs/AlAs multiple quantum well. The modulation was evaluated by measuring the cross-correlation signal which is the second harmonic light generated by the probe pulse reflected from the sample and the gate pulse. The intensity of the correlation-signal decreases owing to the generation of the exciton quantum beat, and recovers with dephasing of the quantum beat oscillation. Moreover, we found that the decrease caused by the quantum beat is larger than that by changing the refractive index due to the exciton generation. These results indicate that quantum beats can be a potential mechanism to enable low power operation of ultrafast optical switches.

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1. Introduction

Exciton quantum beats in nanostructured semiconductors have attracted considerable interest in ultrafast physics [1-12]. In particular, coherent oscillations of the quantum beats provide information regarding wave packet dynamics as well as exciton dephasing time [2-4] and the state of exciton-light coupling [10]. Furthermore, quantum beats have also shown potential properties for application in devices such as terahertz emitters [5] and ultrafast optical switches [9]. In particular, in the case of nanostructured semiconductors, frequency tunable terahertz devices and ultrafast optical switches with controllable response time can be realized by using the quantum beats, because quantum beat period can be controlled by quantum confinement and quantum confined Stark effects.

On the other hand, the most typical mechanism of light modulation for ultrafast optical switches has been based on the change in the refractive index caused by the optical pump-induced change of the carrier density, which is usually observed as changes in transmittance or reflectivity [13-17]. However, a higher density excitation to achieve further faster and larger response causes several problems, such as an increase in the consumption power as well as many-body effects. The use of coupled quantum dot-cavity systems is one realistic approach that can be utilized as a method to decrease in the pump power [18-25]. Then, if the modulation efficiency in quantum structures is improved, a further efficient operation will be expected. Because the quantum beat oscillations are caused by only simultaneous excitation of two exciton states, the ultrafast response can be induced even under weak excitation conditions by using the first cycle of the oscillation. In particular, it is possible to control the exciton energies in nanostructured semiconductors, which means that the response speed is controllable even in simple structures. Therefore, in this work, we demonstrate the modulation effects on the pulse based on the quantum beats. The modulation property was evaluated from the cross-correlation signal. As a result, we show that quantum beats of excitons play an important role in the modulation of the probe pulse. The modulation effect of the probe pulse observed in the cross-correlation signals is discussed from the point of view of quantum beat coherence.

2. Experiment

The sample used in this work was a $(GaAs)_{35}/(AlAs)_{35}$ multiple quantum well (MQW) grown on a (001) GaAs substrate by molecular-beam epitaxy, where the subscripts denote the constitution layer thickness by a monolayer unit of 0.283 nm. The MQW period was 50. The transient response was measured by a time-resolved reflection-type pump-probe technique at 3.4 K. The laser source used was a mode-locked Ti:sapphire pulse laser, delivering approximately 120 fs pulse with the repetition rate of 80 MHz. The laser energy of 1.573 eV almost locates at the center between the heavy-hole (HH) and light-hole (LH) exciton energies [6]. Since the amplitude of the quantum beat reaches

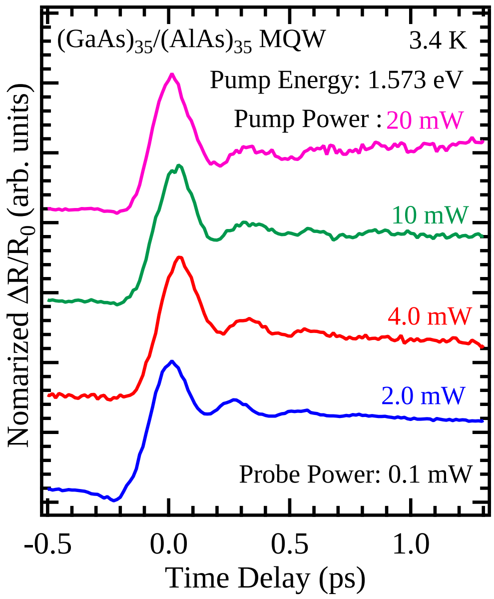

Figure 1. Transient signal measured by the reflection-type pump-probe technique for various pump powers at 3.4 K. The probe power was kept at 0.1 mW.

maximum value at this energy, we considered that the largest modulation effects will be obtained. Considering the relationship between the laser spectral width and the HH-LH exciton splitting energy, the slight deviation of the pump energy is not important in this sample [6]. The pump power was changed from 1.0 to 20 mW, while the probe power was kept at 0.1 mW. Cross-correlation signals were measured to evaluate the pulse modulation effect. Since the intensity of the second harmonic light due to the spatial and temporal overlap between probe pulse reflected from the sample and the gate pulse was recorded as the cross-correlation signal, the signal demonstrates the temporal shape of the probe pulse. The setup was described in detail in our previous report [26].

3. Results and Discussion

Figure 1 shows the time-resolved reflection-type pump-probe signals observed by different pump powers. All signals exhibit the oscillatory structure with the period of 253 fs that corresponds to 16.3 meV of the HH-LH exciton splitting energy in this sample. Therefore, this oscillation arises from the HH-LH exciton quantum beat [6]. While the saturation of the pump-probe signal intensity [27], and the decrease in the signal intensity by Rabi oscillations [28-30] with increasing pump-power have been observed, these effects were not observed in this power region remarkably. In the case of the pump power of 20 mW, while the exciton quantum beat was observed, the oscillation is unclear due to dephasing. Therefore, we measured the cross-correlation signals less than the pump power of 10 mW.

Figure 2(a) depicts the pump-power dependence of the cross-correlation signal.

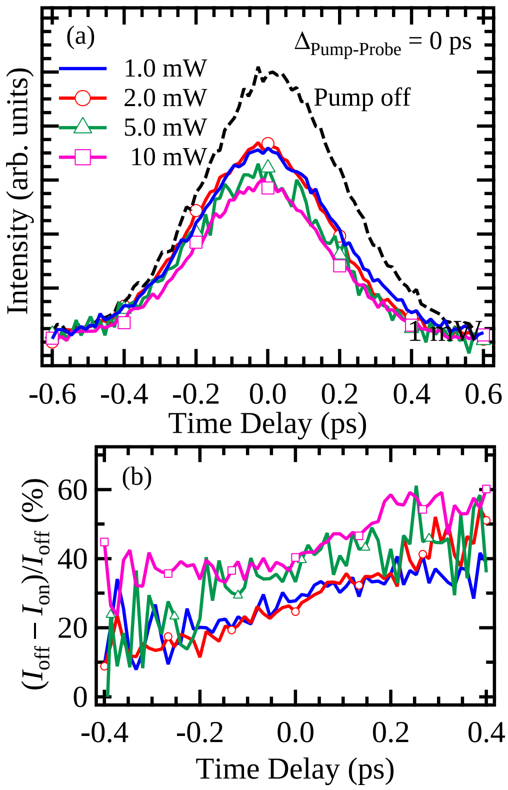

Figure 2. (a) Dependence of cross-correlation signal on pump power at $\Delta_{\text{Pump-Probe}} = 0$ ps. The dotted curve indicates the signal that is obtained in "pump off" condition. (b) Dependence of the intensity ratio on pump power. The symbols used here are the same as in (a).

The dotted curve indicates the signal obtained in "pump off" condition. Time delay between the pump and probe pulses $\Delta_{\text{Pump-Probe}}$ was 0 ps. Signal intensity decreases with increasing pump power. This decrease is attributed to the increase in the number of excitons.

To quantify the amount of modulation, we evaluated the ratio of the signal intensities, $(I_{\text{off}} - I_{\text{on}})/I_{\text{off}}$, where I_{on} and I_{off} indicate the signal intensities obtained in "pump on" and "pump off" conditions, respectively. In Fig. 2 (b), the evaluated results are summarized for an effective time region from -0.4 to 0.4 ps. At the highest excitation power of 10 mW, the amount of the maximum modulation reaches beyond 50% at around 0.2 ps.

To clarify the effect of quantum beat oscillations on the pulse modulation, we measured the cross-correlation signals for different $\Delta_{\text{Pump-Probe}}$, as shown in Fig. 3(a). The pump power was 1.0 mW. The dotted curve indicates the signal obtained by the "pump off" condition. Here, $\Delta_{\text{Pump-Probe}}$ is indicated by using the phase of the quantum beat ocsillation, because quantum beats of HH-LH excitons observed by the pump-probe technique were described as cosine-like oscillations [1]; π is the first dip and 2π is the

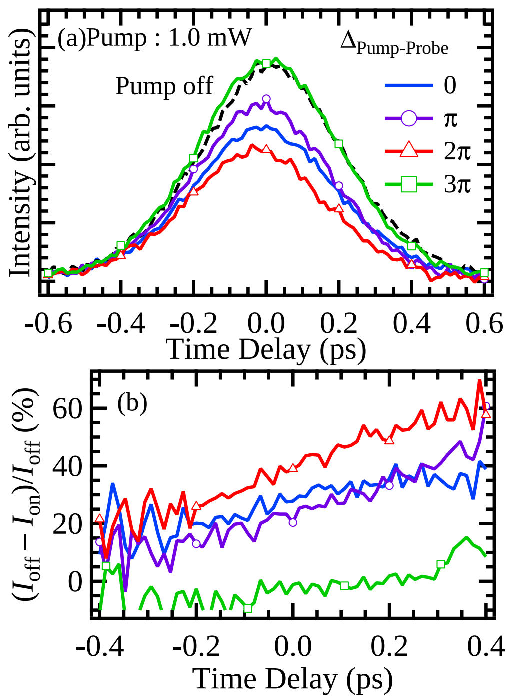

Figure 3. (a) Dependence of cross-correlation signal on $\Delta_{\text{Pump-Probe}}$ for pump power of 1.0 mW. The value of $\Delta_{\text{Pump-Probe}}$ is indicated as the phase of the quantum beat. The dotted curve indicates the signal obtained in "pump off" condition. (b) Dependence of the intensity ratio on $\Delta_{\text{Pump-Probe}}$. The symbols used here are the same as in (a).

next peak, and so on. Signal intensity changes with $\Delta_{\text{Pump-Probe}}$ in a non-monotonic behavior. The fact that the signal at 2π is weaker than the signal at π is crucial point in the present work.

Figure 3(b) summarizes the evaluated ratios of the signal intensities. Interestingly, the modulation reaches a maximum at 2π, as indicated by open triangles in Fig. 3(a). Furthermore, the modulation disappears at 3π, as indicated by open squares in Fig. 3(a). The reflectivity change signal in Fig. 1 shows that the exciton quantum beat disappears at around 3π corresponding to approximately 0.4 ps. While the reflectivity change remains at this time delay, the correlation signal does not show the modulation. Therefore, it was concluded from these results that the coherence of the quantum beat importantly determines the magnitude of the modulation of the pulse.

When the coherence of quantum beat oscillation contributes to the pulse modulation, increasing the pump-power modifies the $\Delta_{\text{Pump-Probe}}$ dependence by dephasing of the quantum beat owing to the exciton-exciton scattering. Figures 4(a), 4(b) and 4(c) show signal intensity ratios that were obtained for various $\Delta_{\text{Pump-Probe}}$, measured at 2.0, 5.0 and 10 mW pump power conditions, respectively. When the intensity at 2π (indicated by open triangles) is focused, the intensity decreases with increasing pump power; a decrease of 20% from 2.0 mW to 10 mW. On the other

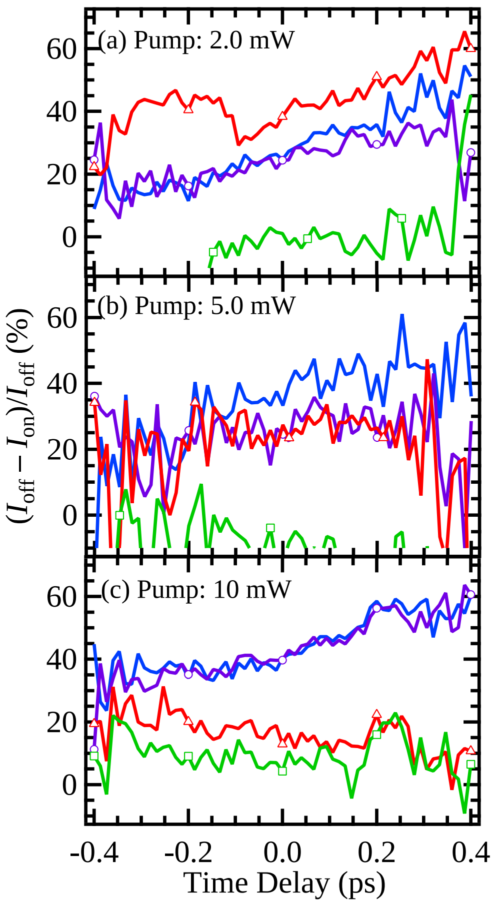

Figure 4. Signal intensity ratios at various $\Delta_{\mathrm{Pump-Probe}}$ measured at (a) 2.0, (b) 5.0, and (c) 10 mW pump power conditions. The symbols used here are the same as in Fig. 3(a); the open circle, open triangle and open square indicates the results obtained at $\Delta_{\mathrm{Pump-Probe}}$ of π, 2π and 3π.

hand, when the intensity at $\Delta_{\mathrm{Pump-Probe}}$ of 0 (depicted by solid curves) is focused, the intensity does not show remarkable change, in particular, from 5 to 10 mW. In the pump-power dependence of the reflectivity change that is shown in Fig. 1, the quantum beat exhibits strong dephasing and weaker oscillation at higher pump power values, which is attributed to the exciton-exciton scattering.

To compare the reflectivity change and the amount of modulation, the values of the signal intensity ratio at time delay of 0 ps that were obtained for various pump conditions were plotted as a function of $\Delta_{\mathrm{Pump-Probe}}$ in Fig. 5. In the case of pump power values smaller than 2.0 mW, the signal intensity ratio reaches the maximum at 2π. However, the signal intensity ratio at 2π decreases with increasing the pump power. The remarkable point is that the value of signal intensity ratio at 2π obtained at the pump powers of 1.0 and 2.0 mW is almost the same as that obtained at 0 by the pump power of 10 mW. This clearly demonstrates that it is possible to use quantum beats for

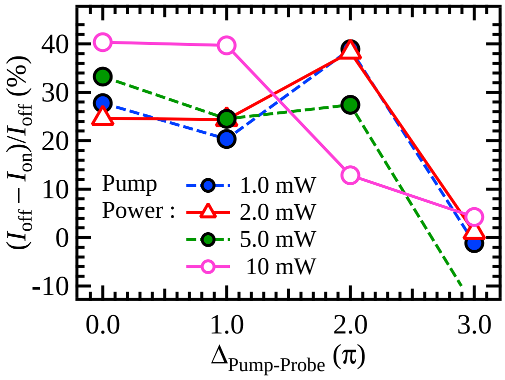

Figure 5. Values of the signal intensity ratio at time delay of 0 ps, obtained for various pump conditions, as in Figs. 4.

low-power operation of the pulse modulation.

Finally, we discuss the mechanism of modulation by quantum beats. If only the reflectivity change due to the exciton generation causes a decrease in the second harmonic signal, the modulation should last after 3π. However, the end of the quantum beat oscillation terminates the modulation; the coherence of the quantum beat is an important factor to modulate the probe pulse. While the reflectivity change continues beyond 1 ps, the intensity of the correlation signal beyond 3π is almost the same as the one that is obtained in "pump off" condition. Therefore, the exciton generation by the pump hardly contributes to the change in correlation signal intensity. Namely, the generation of the quantum beat is the main origin of the change in the correlation signal intensity. Unfrotunately, there is no theoretical reference to explain our results. As a possible explanation, we considered the variation of the reflected light phase, described as $\exp(i\theta)$. Two phase components are created by the generation of the quantum beat; one is the usual reflection component, while another one depends on the quantum beat oscillation. When the quantum beat is generated, the HH exciton and the LH exciton components have different phases, which are usually antiphase [31]. This antiphase oscillation induces the decrease in the correlation signal intensity. On the other hand, after the termination of quantum beat oscillation, there is no antiphase component, so that the intensity recovers. In the case of the time-resolved four-wave-mixing (TR-FWM) signal, the oscillation of the cross-correlation signal by Fano resonance have been reported [32, 33], in which the TR-FWM signal intensity decreases with increasing time delay. Considering this oppsit behavor, the pulse modulation in our result orignates from not variation of the higher order optical nonlinearity but that of the phase the reflected pulse by the pump. At $\Delta_{\text{Pump-Probe}} = 0$, the amplitude of the quantum beat depends on the generated exciton populations, so that the signal intensity ratio almost depends on the pump power. However, the rapid dephasing of the exciton at the higher excitation condition decreases the modulation effect after $\Delta_{\text{Pump-Probe}} = \pi$. This mechanism is proposed as an explanation for pulse modulation.

4. Conclusion

We have investigated the mechanism of pulse modulation that is induced by the quantum beat oscillation, which is an important step towards low power operation of ultrafast optical switches, in a GaAs/AlAs MQW. We found that pulse modulation by quantum beat oscillations, evaluated from the cross-correlation signal, can be larger than the usual refractive index change induced by the exciton generation. As the pump power increases, the amount of the modulation decreases; quantum beat coherence is a key point for the modulation. Our results suggest that using quantum beats for pulse modulation can contribute to the low-power operation of ultrafast optical switches.

Acknowledgments

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