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(Citation)
Journal of Applied Physics,111(2):023505-023505

(Issue Date)
2012-01-15

(Resource Type)
journal article

(Version)
Version of Record

(URL)
https://hdl.handle.net/20.500.14094/90001641

Observation of quantum beat oscillations and ultrafast relaxation of excitons confined in GaAs thin films by controlling probe laser pulses

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(Received 6 September 2011; accepted 21 November 2011; published online 19 January 2012)

We study the quantum beats and relaxation dynamics of exciton center-of-mass motion confined in GaAs thin films by a reflection-type pump-probe technique. By using spectrally narrowed probe pulses with energies comparable with the exciton energy separation, oscillations caused by quantum beats between the confined excitons and ultrafast responses which are shorter than their lifetime appear. This appearance of quantum beats does not result from the so-called detection process. Our results demonstrate that the reduction of the destructive interference of the probe pulse in the sample is a key factor to observe the excitonic quantum beats. © *2012 American Institute of Physics*. [doi:10.1063/1.3676429]

I. INTRODUCTION

Interference between two exciton states, i.e., excitonic quantum beats, in nanostructured semiconductors are an important phenomenon in ultrafast physics,[1–7] and they have been applied in the generation of terahertz electromagnetic waves[8–10] and the development of ultrafast optical switches.[11–13] In quantum well systems, the quantum beats of the heavy-hole (HH) and light-hole (LH) excitons have been observed by a four-wave-mixing (FWM) technique[2,3] and a pump-probe technique.[5,7,14] On the other hand, in the confinement systems of the center-of-mass motion of excitons, although quantum beats have been observed by a FWM technique in CuCl nanocrystals, thin films,[15,16] and GaAs thin films,[11] to our knowledge, the quantum beats have not been measured by a pump-probe technique, which corresponds to the modulation of the exciton population. In particular, realization and observation of the modulation of the exciton population is important in the application of ultrafast optical devices.

In a confinement system of the center-of-mass motion of excitons, several exciton states exist in a narrow energy region because of the small quantum confinement effect. Therefore, excitation of several states by using broadband pulses induces various phenomena, including perturbed free induction decay[17] and the optical Stark shift,[18] which may prevent the generation or observation of the excitonic quantum beat. Here, we focus on the realization of excitonic quantum beats corresponding to the modulation of the exciton population by using probe pulses, in which spectral width and energy are controlled. In this work, we have investigated the dynamics of quantum beats of excitons confined in GaAs thin films by a reflection-type pump-probe technique with controlled probe pulses. As the spectral width of the probe pulses decreases, an ultrafast decay component much faster than the exciton energy relaxation and an oscillatory structure appear, while the transient signal probed by broadband spectral width pulses shows only exciton relaxation. We discuss the origin of the ultrafast decay component and the oscillation in the pump-probe signals from the results of the probe-energy dependence measured using probe pulses with various spectral widths.

II. EXPERIMENT

The sample used in the present study is double heterostructure thin films 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 sufficient thickness to confine the excitons in the GaAs thin films.[11,19–21] The GaAs layer thickness is larger than the exciton Bohr radius (11 nm); therefore, the center-of-mass motion of excitons is confined. The transient response was measured at 4 K by using a reflection-type pump-probe technique. The light source used was a mode-locked Ti:sapphire pulse laser with a pulse width of approximately 200 fs and a repetition rate of 80 MHz. The pump energy was tuned at the exciton resonance of 1.5158 eV. Because the pump and probe densities were kept at 12 and 1.2 nJ/cm^2, respectively, high-density excitation effects were negligible. We varied probe pulses by using a slit placed between the grating pair. By changing the slit width, the spectral width of the probe pulses ΔE was controlled in the range from 0.9 to 20 meV, and the center energy of probe pulses was changed from 1.5158 to 1.5184 eV by changing the slit position. The pump and probe beams were polarized orthogonally to each other, which results in elimination of the pump-beam contribution to the probe beam. The pump beam was chopped at 2 kHz, and the intensity of the reflected probe beam was modulated. The probe intensity detected by a Si photodiode was fed into a lock-in amplifier, which detects the reflectivity change in the probe at 2 kHz. We measured the detection-energy dependence of the reflectivity change by using a monochromator with a resolution of 0.27 nm. The probe light dispersed by the monochromator was detected by an amplified Si

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photodetector. In order to estimate the exciton energies, we also measured the photoluminescence (PL) spectrum by using a semiconductor laser with an energy of 1.84 eV

III. RESULTS AND DISCUSSION

Figure 1(a) shows the PL spectrum for the evaluation of exciton energies. By comparison of their peak energies with the calculation results for exciton energies based on the model of quantization of the center-of-mass motion of excitons,[20,21] we determined the origins of the PL peaks. This calculation does not include the effect of the radiative shift caused by coupling with light fields. The notations $n = X$ HH (LH) indicate the Xth quantized HH (LH) exciton states.

Figure 1(b) shows reflectivity changes probed by various ΔE at 1.5164 eV. Each signal was normalized at the maximum intensity. The signal measured in the negative time region originates from the polariton propagation.[20–22] The signal clearly varies with ΔE of the probe pulse. The signals obtained by using a probe pulse with $\Delta E > 1.2$ meV show ultrafast components around zero delay, which had not been observed by a pump-probe technique in our previous studies, which employed a pump-probe technique with the broadband pulses to probe the response of the several exciton states. In the case of the broadband pump-probe, the lowest exciton showing the strongest response decides the signal profile. On the other hand, the signals obtained by using a probe pulse with $\Delta E = 0.9$ meV clearly show the oscillatory structure.

To clarify the origin of the oscillatory structure and the ultrafast response, we measured the probe-energy dependence of reflectivity changes under the pump condition of $n = 2$ HH exciton, as shown in Fig. 2. In this measurement, a pump pulse with $\Delta E = 20$ meV and probe pulses with $\Delta E = 2.0$ meV were used. The top signal indicates the reflectivity change measured by the probe pulse with $\Delta E = 20$ meV, as a reference. Each signal was normalized at the maximum intensity. The signal probed by the $\Delta E = 2.0$ meV pulse at $n = 2$ HH exciton exhibits a profile similar to that probed by the broadband pulse; this is attributed to the strongest response of the $n = 2$ HH exciton. This strongest response determines the signal profile when the spectrum of the probe pulse sufficiently overlaps the $n = 2$ HH exciton energy, as mentioned above. On the other hand, the signals probed at higher energy demonstrate a relatively different response. In particular, an ultrafast decay component appeared from 1.5164 eV to 1.5168 eV. The relaxation time of this component is approximately 2.0 ps, which is much faster than the energy relaxation time of excitons.

To elucidate the origin of the ultrafast response, the probe-energy dependence of the reflectivity change was measured with a much narrower probe pulse with $\Delta E = 0.9$ meV and a pump pulse with $\Delta E = 20$ meV. As shown in Fig. 3, only the signal probed at 1.5166 eV clearly exhibits the ultrafast decay component. Moreover, oscillatory structures lasting over 30 ps appeared in the signals obtained at 1.5164 eV and 1.5184 eV. The oscillation period of 9.8 ps for the first cycle in the signal at 1.5164 eV corresponds to 0.42 meV, which agrees with the energy separation of 0.41

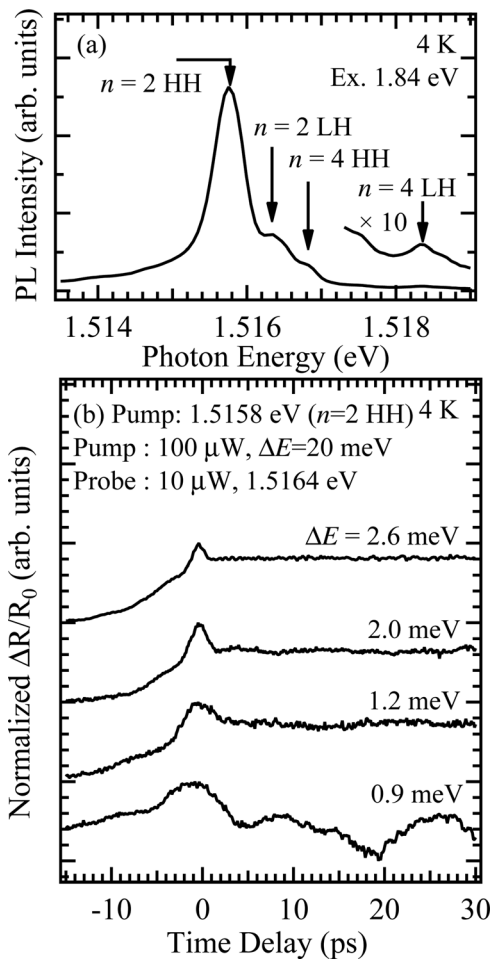

FIG. 1. (a) PL spectrum in GaAs thin films. (b) ΔE dependence of reflectivity-change signal observed at 1.5164 eV. The pump beam was kept at $\Delta E = 20$ meV and the center energy of 1.5158 eV.

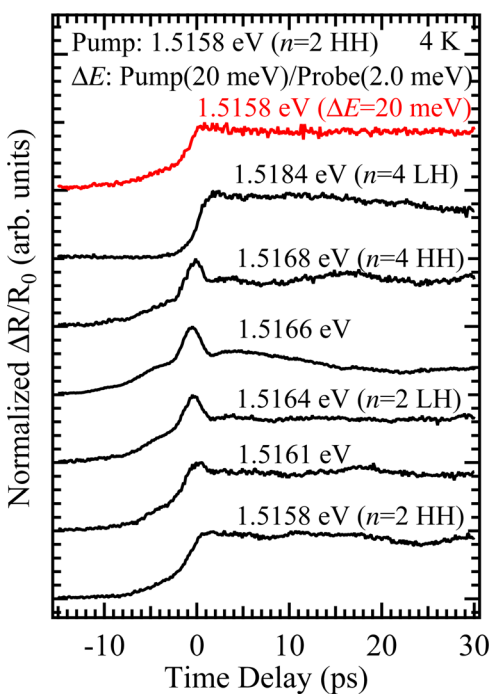

FIG. 2. (Color online) Probe-energy dependence of the reflectivity-change signal probed at pulse with $\Delta E = 2.0$ meV under the exciton resonance of $n = 2$ HH. The top profile was measured by the probe pulse with $\Delta E = 20$ meV.

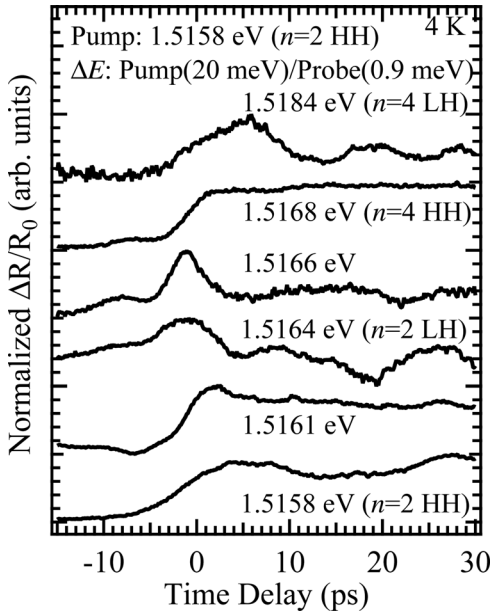

FIG. 3. Reflectivity change signal probed by a pulse with $\Delta E = 0.9$ meV measured at various probe energies.

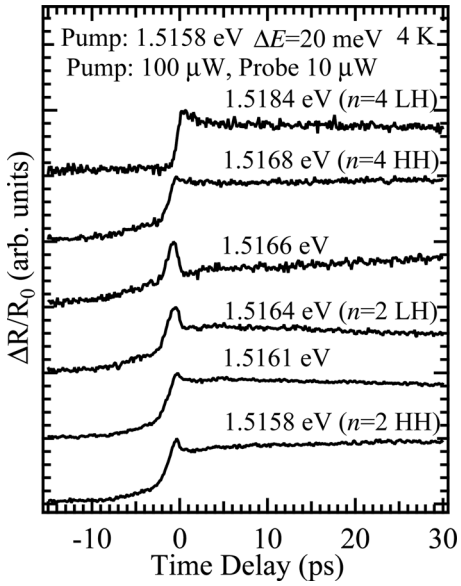

FIG. 4. The detection-energy dependence of reflectivity changes measured using a monochromator. The pump and probe pulses are $\Delta E = 20$ meV. The resolution of the monochromator was 1.0 meV.

meV between the exciton states of $n = 2$ LH and $n = 4$ HH estimated from the PL spectrum. Hence, the oscillatory structure results from the excitonic quantum beat between the $n = 2$ LH and $n = 4$ HH. In addition, the quantum beat between the $n = 6$ HH and $n = 4$ LH (0.4 meV) excitons is observed at 1.5184 eV as the first cycle of the signal. Here, because of the broadening of the $n = 4$ LH state, although the PL from the $n = 6$ HH state barely appears in the PL spectrum, the exciton state exists.

Here, we address the origin of the ultrafast decay component at 1.5166 eV. Because the energy of 1.5166 eV is almost the center energy of the $n = 2$ LH and $n = 4$ HH excitons, the quantum beat oscillation should be observed most clearly;[5] however, the oscillation amplitude is very small. The reason for this small oscillation amplitude is attributed to the momentum relaxation of exciton population from the $n = 4$ HH state to the $n = 2$ HH state. The ultrafast momentum relaxation leads to the disappearance of the oscillatory amplitude.[7,23] The reduction of the population at the exciton state of $n = 4$ HH causes the disappearance of the oscillatory structure. Moreover, in the case of $\Delta E > 1.2$ meV, the overlapping of the strong response due to the $n = 2$ HH exciton on the transient signal leads to the disappearance of the oscillation and ultrafast component.

Furthermore, the oscillatory periods in the signals at 1.5184 eV and 1.5164 eV change with time evolution. In the case of polariton beats, the propagation process changes the oscillatory periods owing to the difference in group velocities.[24–26] Hence, the change in the period results from the propagation of the exciton-polariton confined in the GaAs thin films. In addition, as is well known, the quantum beat oscillations observed by the pump-probe technique are described by the form of $\cos(\Delta E_{ex}t)\exp(-\gamma t)$, where ΔE_{ex} is the energy separation of two exciton states and γ is the dephasing rate originating from the relaxation from the upper level to the lower one.[27,28] Therefore, the observed quantum beats in our measurement indicate the existence of the relaxation between the quantized states of center-of-mass motion.

Finally, we discuss the difference between our method and the so-called detection process.[14] When the superposition of the responses of the several exciton states with different phases prevents the observation of the quantum beats, the detection process was considered as one of the possible reasons for the observation. Thus, we measured the detection-energy dependence of reflectivity changes by using a monochromator with a resolution of 1.0 meV under the pump condition of the $n = 2$ HH exciton. Both the pump and probe pulses have $\Delta E = 20$ meV. Figure 4 shows the reflectivity-change signals observed at various detection energies. If appearance of the excitonic quantum beat is attributed to the detection process, the same results as those shown in Fig. 3 should be obtained. However, the results shown in Fig. 4 are quite different, indicating that the appearance of the quantum beat of the confined excitons results not from the detection process but from the control of the probe pulses before reflection from the sample. The use of broadband probe pulses induces the various polariton modes coherently, so that destructive interference between those polariton beats occurs in the sample during the propagation. Therefore, the reflected probe pulse hardly shows oscillatory structures although only a single exciton component is observed; the coherent oscillation disappears. On the other hand, in the case of spectrally narrow probe pulses, the destructive interference is suppressed, which is an important factor for the appearance of the quantum beats of confined excitons on the transient signals.

IV. CONCLUSION

In summary, we investigated the dynamics of quantum beat oscillation and the transient response of excitons confined in GaAs thin films by changing ΔE of probe pulses. For the control of the transient response, both the spectral width

and tuned energy of the probe pulse play important roles in generating an ultrafast response and quantum beat oscillations of confined excitons. Under the condition that ΔE is equal to the energy separation of two exciton states, which is the condition for confinement of center-of-mass motion of excitons, quantum beats appear in the transient signals. Moreover, the momentum relaxation of the higher exciton states acts as an origin of the ultrafast population relaxation, causing the ultrafast response. By comparing these results with the detection-energy dependence by using a monochromator, we found that the appearance of the quantum beats does not result from the so-called detection process. These results demonstrate that an observation of the quantum beats of confined excitons is realized using probe pulses with controlled spectral width and energy.

ACKNOWLEDGMENTS

This work was partially supported by Grants-in-Aid for Scientific Research (No. 23656050) from the Ministry of Education, Culture, Sports, Science, and Technology (MEXT) of Japan.

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