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Quantum beats of type-I and type-II excitons in an $In_xGa_{1-x}As/GaAs$ strained single quantum well

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We have investigated the quantum beat of the type-I heavy-hole (HH) and the type-II light-hole (LH) excitons in an $In_{0.15}Ga_{0.85}As/GaAs$ strained single quantum well (SQW) using a reflection-type pump-probe technique. The type-II LH exciton consists of the electron and LH located in the $In_{0.15}Ga_{0.85}As$ and GaAs layers, respectively. The energies of the type-I and the type-II excitons were evaluated with photoreflectance spectroscopy. The exciton states were calculated based on a variational method. The time-domain signals clearly show the oscillatory structure with the period corresponding to the splitting energy of the HH and the LH excitons. From the pump-energy dependence of the Fourier transform spectrum and intensity, it is concluded that the oscillation originates from the quantum beat of the type-I HH and the type-II LH excitons. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4748339]

I. INTRODUCTION

Excitonic quantum beats in semiconductor quantum wells (QWs) and superlattices are an important phenomenon from the perspective of understanding ultrafast coherent dynamics of excitons. This phenomenon can be exploited for potential applications in optodevices and in sources of terahertz electromagnetic wave.[1–15] Typical excitonic quantum beats originate from the impulsive interference between the heavy-hole (HH) and light-hole (LH) exciton states in a QW, and the beating period is determined by the HH-LH splitting energy ($\Delta E_{\text{HH-LH}}$). The excitonic quantum beats in type-II superlattices have been observed by photoluminescence dynamics under magnetic fields.[6] However, the quantum beat between the type-I and the type-II excitons has not yet been reported. The magnitude of the polarization of charge oscillation due to the HH-LH quantum beat depend on the intersubband transition dipole moment described by $\langle \text{LH}|x|\text{HH}\rangle$,[5] so that the spatial separation of the positions of the LH and HH induces the large polarization. In the case of the quantum beat of the type-I and the type-II excitons, therefore, it is expected that the spatial separation of the relevant states leads to generate intense emission of terahertz electromagnetic waves because of the large polarization.

In the present paper, we report on the quantum beat of the type-I HH and the type-II LH excitons measured using a reflection-type pump-probe technique in a $GaAs/In_{0.15}Ga_{0.85}As/GaAs$ strained single quantum well (SQW). The period of the oscillatory structures observed in the time-domain signals corresponds to $\Delta E_{\text{HH-LH}}$. In this sample, the HH exciton confined in the $In_{0.15}Ga_{0.85}As$ well layer is type-I, and the LH exciton consisting of the LH in the GaAs barrier layer and the electron in the $In_{0.15}Ga_{0.85}As$ well layer is type-II. We discuss the quantum beat of the type-I HH and the type-II LH excitons from aspects of the pump-energy dependence and dephasing process.

II. EXPERIMENT

The sample used is a $GaAs/In_{0.15}Ga_{0.85}As/GaAs$ strained SQW grown on a (001) GaAs substrate by molecular-beam epitaxy. The thickness of $In_{0.15}Ga_{0.85}As$ layer is 3 nm and the thickness of the GaAs layer is 100 nm.[16] Under a pseudomorphic growth condition on a GaAs substrate, a compressive strain in the $In_{0.15}Ga_{0.85}As$ layer due to the lattice mismatch lifts the degeneracy of the HH and the LH bands at the Γ point. It has been confirmed that the strain effect results in two types of potential configurations in $In_xGa_{1-x}As/GaAs$ strained QWs with the indium concentration ranging from 0.1 to 0.3:[16–19] the type-I potential for the HH band and the type-II potential for the LH band, as shown in the inset of Fig. 1. In a $GaAs/In_{0.15}Ga_{0.85}As/GaAs$ strained SQW, the electron and HH are confined in the $In_{0.15}Ga_{0.85}As$ layer, whereas there is no potential confinement for the LH envelope function located in the thick GaAs layer. Though the LH state in the SQW is unconfined, the type-II transition is possible because of the excitonic effect that attracts the unconfined LH envelope function via the Coulomb potential of the electron confined in the $In_{0.15}Ga_{0.85}As$ layer.[16,19] This is a key point in this work.

The time-domain signals were measured using a time-resolved reflection-type pump-probe technique at 10 K. The laser source was a mode-locked Ti:sapphire pulse laser delivering a pulse with a pulse width of about 100 fs at a

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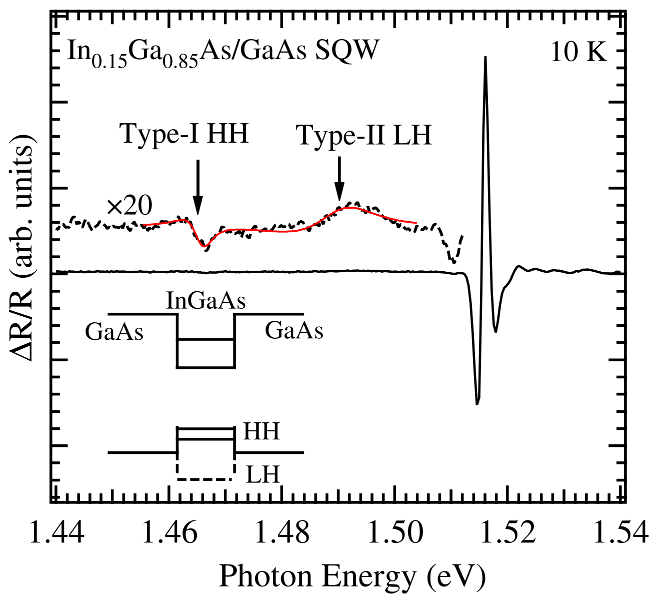

FIG. 1. PR spectrum of the $In_{0.15}Ga_{0.85}As/GaAs$ strained SQW sample at 10 K. The solid curve in the magnified spectrum is the fitted PR spectrum. The arrows indicate the excitonic transition energies. The inset shows a schematic diagram of the band configuration of the sample.

repetition rate of 80 MHz. The pump and probe beams were orthogonally polarized to each other in order to eliminate the pump-beam contribution to the probe beam. The pump energy was changed from 1.46 to 1.50 eV in the exciton-energy region. The pump and probe densities were kept at 120 and 12 nJ/cm^2, respectively. The exciton energies in the $In_{0.15}Ga_{0.85}As/GaAs$ strained SQW were evaluated from a photoreflectance (PR) spectrum at 10 K. The probe light for the PR measurement was produced by combination of a 100 W-halogen lamp and a 32-cm single monochromator with a resolution of 0.4 nm. The modulation light was the 632.8 nm line of a He-Ne laser chopped at 630 Hz. The reflected light was detected using a Si photodiode, and the modulated signal was measured using a conventional lock-in technique.

III. RESULTS AND DISCUSSION

At first, we characterize the energies of the type-I and the type-II excitons. The PR spectrum of the sample at 10 K is depicted in Fig. 1. To estimate the exciton energies, we performed line-shape analysis based on a first derivative functional form, which is adequate to analyze the excitonic transition energy.[20] The solid curve in the magnified spectrum indicates the fitted result in the exciton-energy region. The arrows show the transition energies obtained from the fitting. We determine the origins of the exciton transitions by comparing these results with the theoretical results.[16] The transition energies of the type-I HH and the type-II LH excitons evaluated using the above-mentioned fitting method are 1.465 and 1.490 eV, respectively. Therefore, $\Delta E_{\text{HH-LH}}$ in the sample is 25 meV.

To generate the quantum beat, the overlap of the envelope functions of the HH and the LH is an important factor. Then, we calculated the envelope functions of the excitons by using a flexible calculation method which is applicable to both the type-I and the type-II excitons.[21] The calculated results are shown in Figs. 2(a) and 2(b) for the type-I HH and the type-II LH excitons, respectively. This method fundamentally involves the expansion of the envelope function of the exciton into a set of Gaussian-type functions; namely, a fixed trial function is not used. It is evident from Fig. 2(b) that the Coulomb potential of the electron in the $In_{0.15}Ga_{0.85}As$ layer attracts the envelope function of the unconfined LH envelope function located in the GaAs layer. As shown in Fig. 2(b), the LH envelope function tunnels into the $In_{0.15}Ga_{0.85}As$ layer; therefore, the overlap between the HH and the LH envelope functions is enough to generate the quantum beat. In the calculation, the binding energies of the type-I HH and the type-II LH excitons are evaluated as 12.3 and 2.7 meV, respectively.

Figure 3 shows the time-resolved reflectivity-changes of the $In_{0.15}Ga_{0.85}As/GaAs$ strained SQW sample measured at various pump energies between the HH and the LH excitons. All the time-domain signals consist of the following two components: a large reflectivity change around 0 ps and an oscillatory structure in the time region less than 1.0 ps. The initial part around 0 ps is attributed to a change in the exciton density in the SQW.[22] The period of the oscillatory structure within 1.0 ps is 145 fs at all pump energies. Moreover, the oscillation amplitude of this structure depends on the pump energy.

To clarify the origin of the oscillatory structure, we performed time-partitioning Fourier transform (FT) for the time-domain signals in the time region from 0.02 to 1.5 ps. The FT spectra at various pump energies are shown in Fig. 4. The FT spectra have a peak at 6.8 THz (28 meV), which almost corresponds to $\Delta E_{\text{HH-LH}}$ of 25 meV. Therefore, the oscillatory structure within 1.0 ps results from the quantum beat of the type-I HH and the type-II LH excitons.

To confirm that the oscillation in the time region within 1.0 ps results from the excitonic quantum beat of the type-I HH and the type-II LH excitons, the integrated intensity of the FT band is plotted as a function of pump energy in

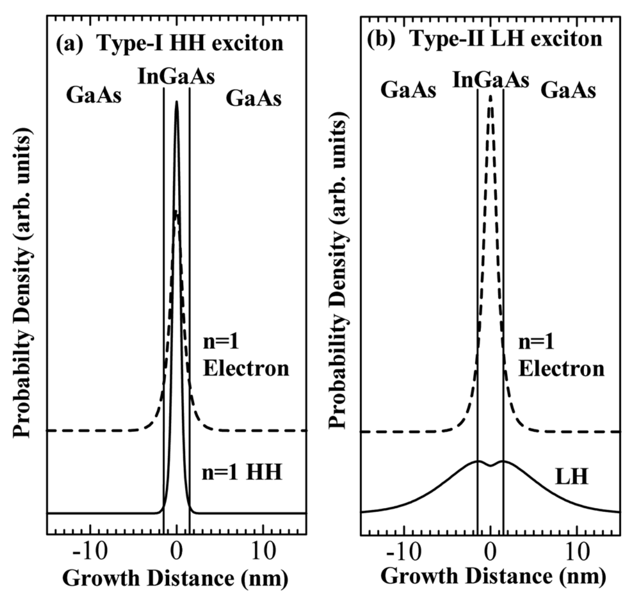

FIG. 2. Calculated results for the probability densities of the electrons, HH, and LH of (a) the type-I HH exciton and (b) the type-II LH exciton in the $In_{0.15}Ga_{0.85}As/GaAs$ strained SQW structure.

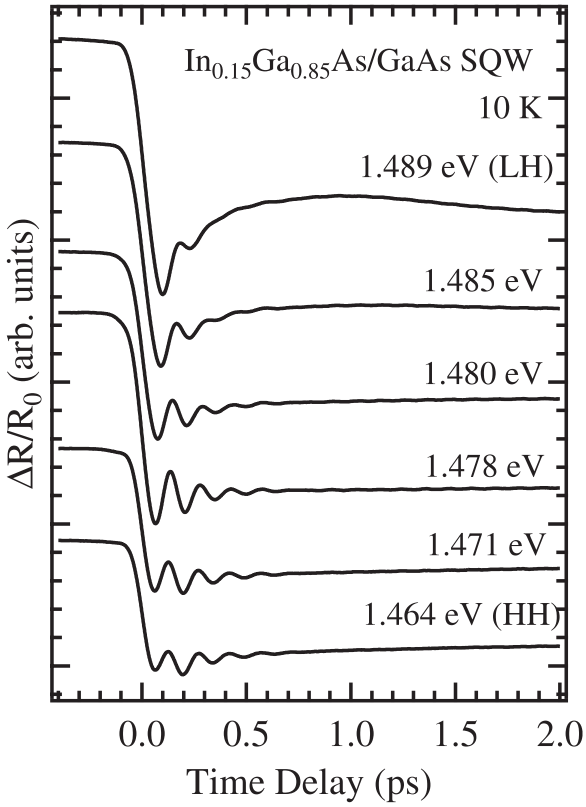

FIG. 3. Pump-energy dependence of the time-domain reflectivity change signals in the $In_{0.15}Ga_{0.85}As/GaAs$ strained SQW.

Fig. 5. The vertical dashed lines indicate the type-I HH and the type-II LH exciton energies evaluated from the PR spectrum. The FT intensity reaches a peak at 1.478 eV around the center energy between the HH and the LH exciton. It is a well-known feature that the intensity of the quantum beat of

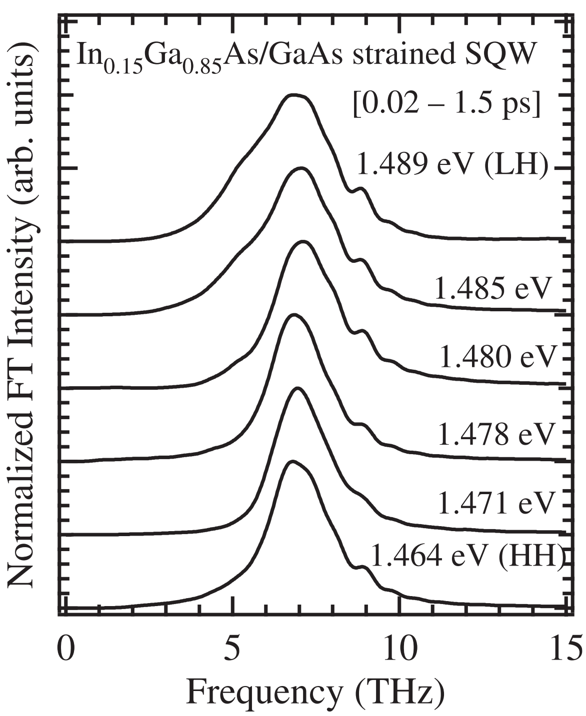

FIG. 4. Pump-energy dependence of the FT spectrum of the time-domain signal in the time region from 0.02 to 1.5 ps.

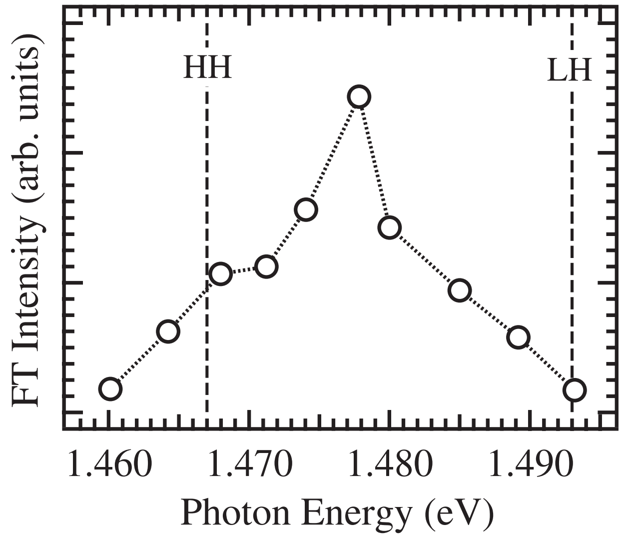

FIG. 5. Pump-energy dependence of the FT intensities of the excitonic quantum beat. The vertical dashed lines indicate the exciton energies.

the HH and the LH excitons essentially reaches a maximum at the center energy between the HH and the LH excitons.[7,8,10] Thus, the result of the pump-energy dependence clearly demonstrates that the oscillatory structure in the time region within 1.0 ps results from the quantum beat of the type-I HH and the type-II LH excitons. The pump-energy dependence of the FT intensity exhibits an asymmetric profile: The weight of the type-I HH exciton is larger than that of the type-II LH exciton. This is due to the fact that the broadening factor of the type-II LH exciton is about 3 times larger than that of the type-I HH exciton as shown in Fig. 1.

The quantum beat oscillation observed by a pump-probe technique is described by a form of $\cos(\omega_{12}t+\phi)\exp(-\gamma_{12}t)$, where ω_{12} is the energy separation of two levels in the frequency unit, ϕ is the oscillation phase, and γ_{12} is the dephasing rate arising from the transition between the two levels.[3,4] In the case of the HH-LH quantum beats, γ_{12} originates from the transition from the LH to the HH states, because the energy of the LH state is larger than that of the HH state. Figure 6 shows the fitting result with the damped harmonic

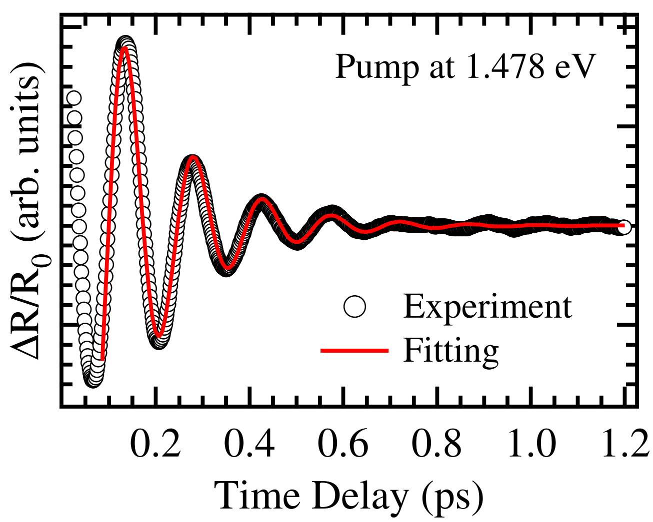

FIG. 6. Fitting result (solid curve) with a damped harmonic oscillation for the quantum beat oscillation, where the open circles indicate the experimental result.

oscillation model for the quantum beat signal at the pump energy of 1.478 eV, where the open circles indicate the experimental result, and the solid curve depicts the fitting result. The oscillation decay time is evaluated as 154 fs, which indicates the dephasing time resulting from the relaxation from the type-II LH subband to the type-I HH subband. This decay time is shorter than that of the quantum beat at Γ point in quantum well and longer than that at π point.[14] Since the quantum beat between the type-I and the type-II excitons includes the real space transfer of the HH and the LH the interfaces of the well and barrier layers considerably affect the decay time, which is quite different from the usual excitonic quantum beats in which all the relevant states are confined in the same well layer. Therefore, scattering by the interface roughness leads to shortening the dephasing time of the quantum beat between the type-I and the type-II excitons.

IV. CONCLUSION

The quantum beat of the type-I HH and the type-II LH excitons in an $In_{0.15}Ga_{0.85}As/GaAs$ strained SQW has been investigated using a reflection-type pump-probe technique. The energies of the type-I HH and the type-II LH excitons were evaluated with PR spectroscopy. The calculated results of the envelope functions indicate that the spatial overlap between the type-I HH and the type-II LH states are sufficient to produce the quantum beat. The time-domain signals clearly show the oscillatory structure with the period corresponding to $\Delta E_{\text{HH-LH}}$. From the pump-energy dependence of the FT spectra and the FT intensity, we have revealed that the origin of the oscillations is the quantum beat of the type-I HH and the type-II LH excitons.

ACKNOWLEDGMENTS

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