PHYSICAL REVIEW B **91**, 125307 (2015)

Rapid dephasing related to intersubband transitions induced by exciton quantum beats observed by a pump-probe technique in a GaAs/AlAs multiple quantum well

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(Received 24 March 2014; revised manuscript received 27 February 2015; published 20 March 2015)

We discuss the dephasing of quantum beat oscillation related to the intersubband transition between heavy-hole and light-hole excitons in a GaAs/AlAs multiple quantum well as measured by a pump-probe technique. We investigate the dependence of dephasing on temperature and pump energy. The analysis of the time-domain signals reveals that a strong quantum beat induces the rapid dephasing related to intersubband transition; the dependencies of the dephasing rate correspond to those of the amplitude. This implies that a quantum beat is useful in applications utilizing an ultrafast optical response.

DOI: 10.1103/PhysRevB.91.125307 PACS number(s): 78.47.jm, 78.47.jg, 78.55.Cr

I. INTRODUCTION

Oscillation by exciton quantum beats in nanostructured semiconductors is an important coherent phenomenon in ultrafast physics [1–19]. In particular, coherent interference between two polarizations generating the quantum beat, e.g., heavy-hole (HH) and light-hole (LH) excitons, is important for applications in devices such as ultrafast optical switches [13,19] and frequency-tunable emitters of terahertz electromagnetic waves [5,8].

Utilization of saturable absorption in quantum dots has been proposed as an approach toward creating ultrafast all-optical switches [20–26]. In a previous study, we reported the possibility of a low-power operation using an exciton quantum beat in a GaAs/AlAs quantum well [19], which is a different approach from the use of the changes in refractive index. While a quantum beat enables the generation of an ultrafast and intense signal based on the optical nonlinearity of excitons [13], ultrafast dephasing of the oscillation is also required. In general, a quantum beat appearing in a pump-probe signal is described as follows:

$$I_{\mathrm{PP}}(t) \propto |\mu_{\mathrm{HH}}\mu_{\mathrm{LH}}|^2[1+\cos(-\omega_{\mathrm{QB}}t)]\exp(-\gamma_{\mathrm{LH-HH}}t), \quad (1)$$

where μ_{HH} and μ_{LH} are the self-dipole moments of the HH and LH excitons, respectively, ω_{QB} corresponds to the HH-LH splitting energy and is the quantum beat frequency, t is the time delay, and $\gamma_{\mathrm{LH-HH}}$ is the dephasing rate caused by the intersubband transition between the LH exciton state and the HH exciton state [1,3,4]. If the intersubband transition is only caused by the scattering from the thermal phonon, the dephasing rate of the quantum beat simply depends on the temperature. Therefore, to enable their use in the ultrafast response applications, in this study, we discuss the dephasing of quantum beat oscillations in pump-probe signals based on the results of the dependence on temperature and pump energy. We find that the dephasing rate depends not on the temperature but rather on the amplitude; it is the strong oscillation that induces rapid dephasing. Such ultrafast dephasing related to intersubband transition enables quantum beats to be used for ultrafast all-optical switching in the next-generation ultrafast optical communication networks.

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II. EXPERIMENT

The sample used in this study was a $(\mathrm{GaAs})_{35}/(\mathrm{AlAs})_{35}$ multiple quantum well (MQW) grown on a (001) GaAs substrate by molecular-beam epitaxy, where the subscripts denote the number of the 0.283-nm-thick monolayers. The MQW period was 50. The quantum beat was measured by a time-resolved reflection-type pump-probe technique. The measurement temperature was changed from 4 K to 180 K. The laser source used was a mode-locked Ti:sapphire pulse laser, delivering an approximately 150-fs pulse with a repetition rate of 80 MHz. The pump and probe powers were kept at 1.20 and 0.06 $\mu\mathrm{J/cm^2}$, respectively. While the pump-induced dephasing of the quantum beat was observed at 1.20 $\mu\mathrm{J/cm^2}$ to some degree in our previous study [19], we chose this pump power to clearly observe the oscillation at higher temperatures. The pump and probe beams were polarized orthogonally to each other to eliminate the contribution of the pump to the probe pulses. The pump beam was chopped at 2 kHz, and then the intensity of the reflected probe beam was modulated. The probe intensity detected by a Si photodiode was amplified by a lock-in amplifier. The exciton energy and line widths were evaluated from the photoluminescence (PL) spectrum at each temperature. The excitation source was a semiconductor laser with a photon energy of 1.85 eV (670 nm) and an excitation intensity of 0.7 $\mathrm{kW/cm^2}$. The emitted light was dispersed by a 32-cm single monochromator with a resolution of 0.3 nm and detected by a liquid-nitrogen-cooled InGaAs-photodiode array.

III. RESULTS AND DISCUSSION

Figure 1 shows the time-resolved reflection-type pump-probe signals observed at the different temperatures. The laser energy was tuned to the almost center energy between the HH and LH excitons at each temperature. The exciton energies were estimated from the PL spectrum. The signal inversion observed above 100 K originates from several factors, including the pump energy and the spectral width of

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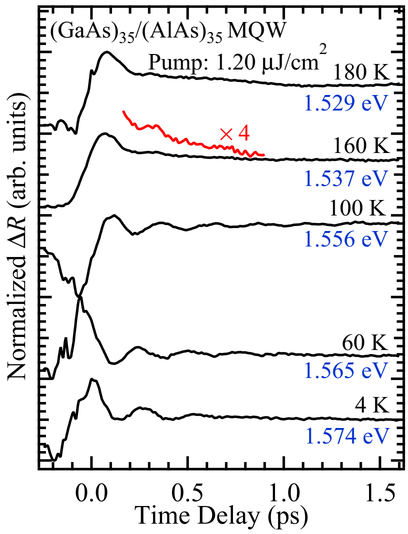

FIG. 1. (Color online) The time-resolved reflection-type pump-probe signals observed at different temperatures. The pump energy was tuned to the center energy between the HH and LH excitons at each temperature.

the laser leading to the phase shift of the oscillation [27]. The inversion itself is not essential for the observation of the quantum beat oscillation. Except for 180 K, the signal shows the oscillatory structure due to the HH-LH exciton quantum beat with the period corresponding to the HH-LH splitting energy in the sample. The amplitude of the quantum beat seems to change with temperature and vanishes at 180 K.

To clarify the change in the oscillation characteristics with increasing temperature, we used a function with three components based on Eq. (1) to analyze the signals:

$$I_{\mathrm{PP}}(t) = C_1[1 + \cos(\omega_{\mathrm{QB}} t + \phi)] \exp(-\gamma_{\mathrm{LH-HH}} t) + C_2 \exp(-\gamma_{\mathrm{HH}} t) + C_3 \exp(-\gamma_{\mathrm{LH}} t), \quad (2)$$

where the first, second, and third terms relate to the quantum beat oscillation and the HH and LH exciton dynamics, respectively. C_1, C_2, and C_3 are the amplitudes of the components, ϕ is the phase of the quantum beat oscillation, and γ_{HH} and γ_{LH} are the damping factors for each component. In Fig. 2, the fitting results for the signal at 4 K, 100 K, and 120 K are shown, with the thick and the thin curves indicating the measured and the fitted results, respectively.

The evaluated dephasing rate of the quantum beat $\gamma_{\mathrm{LH-HH}}$ as a function of temperature is shown by the closed circles in Fig. 3. Interestingly, $\gamma_{\mathrm{LH-HH}}$ does not show a drastic increase with increasing temperature at temperatures less than 100 K and shows a strong increase above 100 K. Furthermore, C_1 does not decrease with increasing temperature and shows a peak at 140 K, as depicted by the open circles. If scattering by the thermally excited acoustic phonons is the main cause of the intersubband transition, $\gamma_{\mathrm{LH-HH}}$ should increase with temperature, as indicated by the dotted line, which was calculated using the Boltzmann constant k_B, temperature T, and Planck's constant h. However, the temperature dependence of $\gamma_{\mathrm{LH-HH}}$ follows that of C_1. Hence, at least in this temperature region, the dephasing of the quantum beat is strongly related to the amplitude. This is the key result in this study.

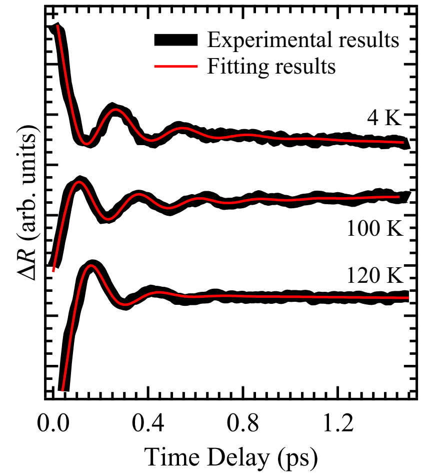

FIG. 2. (Color online) Fitted results for the signal observed at 4 K, 100 K, and 120 K. Thick and thin curves indicate the experimental and the fitted results, respectively.

Moreover, the temperature dependence of C_1 shows a secondary peak at 80 K. The phase of the quantum beat evaluated by Eq. (2) is plotted as a function of temperature in the inset of Fig. 3. The phase is indicated as the value relative to that at 4 K. At 80 K, the phase is clearly inverted. Therefore, the peak at 80 K originates from the phase inversion.

To show that the strong oscillation induces rapid dephasing, we measured the pump-energy dependence of the pump-probe signal at 120 K, as shown in Fig. 4(a). We chose 120 K because the oscillation was relatively clear in comparison with 160 K. The damping rate and amplitude analyzed using Eq. (2) are plotted as functions of the pump energy in Fig. 4(b). Both $\gamma_{\mathrm{LH-HH}}$ and C_1 show the peak at around the center energy of the HH and LH excitons. If the intersubband transition is caused by thermal phonons, this rate should be constant.

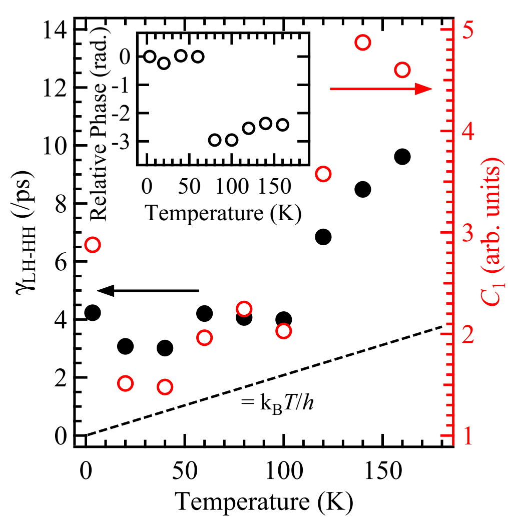

FIG. 3. (Color online) Temperature dependencies of the damping rate $\gamma_{\mathrm{LH-HH}}$ (solid circles) and the oscillation amplitude C_1 (open circles). The dotted curve indicates the scattering rate calculated from the thermal energy. The relationship between the relative phase and the temperature is shown in the inset.

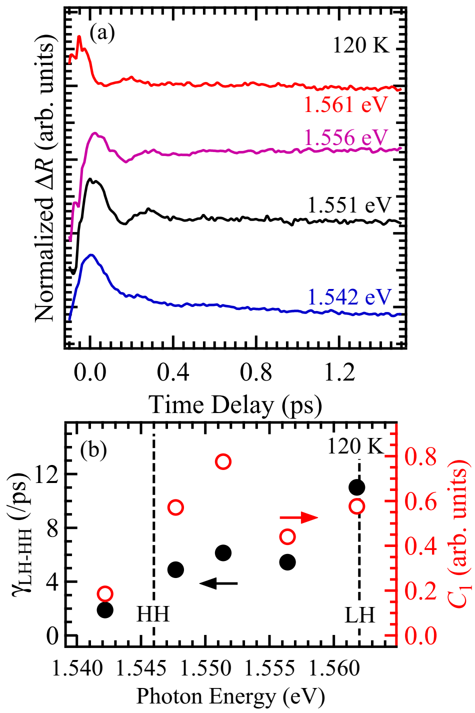

FIG. 4. (Color online) (a) Pump-energy dependence of the reflectivity change signal observed at 120 K. (b) Pump-energy dependencies of the damping rate $\gamma_{\mathrm{LH-HH}}$ (solid circles) and the amplitude C_1 (open circles) of the oscillation at 120 K. Dotted lines indicate the HH and LH exciton energies.

Therefore, we conclude that quantum beat oscillation induces the ultrafast dephasing related to intersubband transition that arises from the simultaneous excitation of two exciton states. In addition, $\gamma_{\mathrm{HH-LH}}$ increases at 1.561 eV. Since the spectral width of the laser is broader than the exciton line width, the free carriers are generated. Therefore, this large dephasing may be caused by the interaction of the excitons with them.

Finally, to discuss the effects of the thermal broadening of the exciton line shapes, we focus on the broadening factors of excitons. We evaluated the homogeneous and inhomogeneous broadening widths obtained from the PL spectra using the convolution of two line shapes described by the Lorentzian and Gaussian functions [28,29]. The PL spectrum at 120 K is shown in Fig. 5. In general, an exciton transition is described by the Lorentzian function

$$L_n(E) \propto \frac{1}{(E-E_n)^2+\Gamma_n^2}, \tag{3}$$

where E_n and Γ_n indicate the exciton energy and the broadening factor that is associated with the homogeneous broadening, respectively. The subscript n refers to either the HH or LH excitons. On the other hand, the inhomogeneous broadening originating from the fluctuation of the well width due to the randomness in the monolayer unit is described by the Gaussian function

$$G_n(E) \propto \exp\left(-\frac{(E-E_n)^2}{2\sigma_n^2}\right), \tag{4}$$

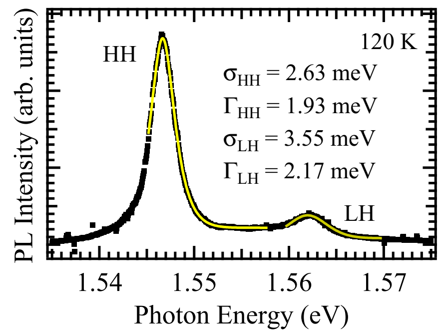

FIG. 5. (Color online) The PL spectrum observed at 120 K and the fitting results by Eq. (6) indicated by dots and solid curves, respectively.

where σ_n^2 indicates the dispersion related to the inhomogeneous broadening. The Voigt function is given by the convolution of these functions:

$$I_{\mathrm{PL},n}(E) \propto \int L_n(E)G_n(E-E')\mathrm{d}E'. \tag{5}$$

The explicit form of the function used for fitting of the data is shown as follows:

$$I_{\mathrm{PL}}(E) \propto \frac{\mathrm{Erf}\left(\frac{-[(E-E_n)+E']}{\sqrt{2}\sigma_n}\right)}{2\left[(E-E_n)^2+\Gamma_n^2\right]} - \frac{\mathrm{Erf}\left(\frac{-[(E-E_n)-E']}{\sqrt{2}\sigma_n}\right)}{2\left[(E-E_n)^2+\Gamma_n^2\right]}. \tag{6}$$

Fitting results for the HH-exciton and LH-exciton lines at 120 K are shown in Fig. 5 by solid curves. The values of the broadening factors obtained by this fitting are also presented. The relationship between Γ_n and the dephasing time T_2 is expressed by $\Gamma_n = 2\hbar/T_2$ [30], where $\hbar$ is Dirac's constant. The exciton dephasing rate estimated from Γ_n of 2 meV is 1.5 ps^{-1}, which is a value smaller than that of the quantum beat. Therefore, this result shows that the generation of the quantum beat induces the rapid dephasing related to the intersubband transition.

IV. CONCLUSION

We have investigated the dephasing of the quantum beat oscillations in a GaAs/AlAs MQW as observed by the pump-probe technique. The quantum beat amplitude depends on the temperature, with the peak value obtained at 140 K. Examination of the dephasing rate and the quantum beat oscillation amplitude for different pump energies and temperatures showed that the dependencies of the dephasing rate on the temperature and the pump energy follow those of the quantum beat oscillation amplitude. These results indicate that the quantum beat induces the rapid dephasing of the oscillation related to the intersubband transition.

ACKNOWLEDGMENTS

This work was partially supported by a Grant-in-Aid for Scientific Research (Grant No. 26289088) from the Ministry of Education, Culture, Sports, Science and Technology of Japan; Hyogo Science and Technology Association; Kansai Research Foundation for Technology Promotion; and the Ogasawara Foundation for the Promotion of Science and Technology.

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