Coupling of coherent longitudinal optical phonons to excitonic quantum beats in GaAs/AlAs multiple quantum wells

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We have investigated the dynamical properties of the coherent oscillations due to the GaAs-like longitudinal optical (LO) phonon and the quantum beat of excitons in two samples of GaAs/AlAs multiple quantum wells with different splitting energies of the heavy-hole and light-hole excitons. The coherent oscillations were measured with a reflection-type pump-probe technique. In the multiple quantum well with the splitting energy almost equal to the energy of the GaAs-like LO phonon, the intensity of the coherent LO phonon are markedly enhanced in comparison with that of the other sample, and the pump-energy dependence of the intensity of the coherent GaAs-like LO phonon shows similar profile to that of the excitonic quantum beat. Moreover, the intensity of the coherent GaAs-like LO phonon is linearly increased with an increase in pump power, which is contrary to a usual generation mechanism, i.e., the displacive excitation of coherent phonon leading to a square dependence. These facts indicate that the excitonic quantum beat acts as a driving force for the coherent GaAs-like LO phonon in the case that the splitting energy of the heavy-hole and light-hole excitons is almost equal to the LO phonon energy.

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I. INTRODUCTION

Coherent phonons generated by ultrashort laser pulses have attracted much attention from the viewpoint of the phonon dynamics in nonequilibrium states.[1–4] Recently, coherent phonons coupled with other coherent oscillations due to photoexcitation, e.g., Bloch oscillations and coherent plasmon, have been investigated in superlattices (SL's) and multiple quantum wells (MQW's).[5–8] Dekorsy *et al.* reported on the coupling between the Bloch oscillation and the coherent longitudinal optical (LO) phonon in GaAs/AlGaAs SL's, and observed an enhancement of the coherent LO phonon oscillations by tuning the frequency of the Bloch oscillation to that of the coherent LO phonon.[5] The enhancement of coherent LO phonons is correlated with the longitudinal polarization along the SL growth direction due to the Bloch oscillation producing terahertz radiation. Quantum beats of excitons in MQW's were studied considerably from an aspect of terahertz radiation.[2,9–11] It was revealed that terahertz radiation from the excitonic quantum beat arises from the longitudinal polarization due to a quantum-confined Stark effect and band mixing between the heavy-hole(HH) and light-hole(LH) states. Thus, we expect the enhancement of the coherent LO phonons by using the longitudinal polarization due to the excitonic quantum beat under the condition that the energy of the excitonic quantum beat is tuned to that of the coherent LO phonon. To our knowledge, however, there is no such report on the enhancement of coherent LO phonons. In SL's and MQW's, the electronic and optical properties are controllable by changing the structural parameters. It is especially possible to control the energies of the HH and LH excitons by using quantum size effects. When the splitting energy of the HH and LH excitons (ΔE_{HH-LH}) in a MQW is tuned to the LO phonon energy (E_{LO}) in the well layer, the excitonic quantum beat may drive the coherent LO phonon via the longitudinal polarization.

In the present paper, we report on the dynamical properties of the coherent GaAs-like LO phonon and the quantum beat of the HH and LH excitons measured by a reflection-type pump-probe technique in two samples of GaAs/AlAs MQW's with different splitting energies ΔE_{HH-LH}. One of the MQW's has ΔE_{HH-LH} almost equal to E_{LO} of GaAs, and the other has ΔE_{HH-LH} much smaller than E_{LO}. We have found that the intensity of the coherent GaAs-like LO phonon is markedly enhanced in the sample with $\Delta E_{HH-LH} \simeq E_{LO}$ and that the resonance profile of the coherent GaAs-like LO phonon is similar to that of the excitonic quantum beat, which is different from that in the other sample with $\Delta E_{HH-LH} \ll E_{LO}$. In addition, the pump-power dependence of the intensity of the coherent GaAs-like LO phonon in the two samples shows remarkable difference. In the sample with $\Delta E_{HH-LH} \simeq E_{LO}$, the intensity of the coherent LO phonon is linearly increased with an increase in pump power, which is contrary to usual generation mechanisms, i.e., the displacive excitation of coherent phonon (DECP) mechanism and the impulsive stimulated Raman scattering (ISRS) mechanism leading to a square dependence.[12] We discuss the difference of the properties of the coherent GaAs-like LO phonon in the two samples from the viewpoint of the coupling of the coherent LO phonon to the excitonic quantum beat.

II. EXPERIMENT

The samples used in the present work are two $(GaAs)_m/(AlAs)_m$ MQW's with $m = 18$ and 35, grown on a (001) GaAs substrate by molecular-beam epitaxy, where the subscript m denotes the thicknesses of the constituent layers in monolayer units (0.283 nm). The total thickness in each MQW is about 600 nm. Hereafter, we will call these samples "(18,18) MQW" or "(35,35) MQW." The HH and LH exci-

TABLE I. HH and LH exciton energies and the splitting energy (ΔE_{HH-LH}) at 10 K in the (18,18) and (35,35) MQW's measured with PLE spectroscopy.

Sample	HH exciton energy (eV)	LH exciton energy (eV)	Splitting energy (meV)
(18, 18) MQW	1.672	1.710	38
(35, 35) MQW	1.565	1.580	15

ton energies and ΔE_{HH-LH} in each sample at 10 K are listed in Table I. These exciton energies were determined by photoluminescence excitation (PLE) spectroscopy. It is noted that ΔE_{HH-LH} of the (18,18) MQW is almost equal to E_{LO} of GaAs that is 36.5 meV.

The quantum beat of the HH and LH excitons and the coherent GaAs-like LO phonon were measured by a reflection-type pump-probe technique at 10 K. The laser source was a mode-locked Ti:sapphire pulse laser delivering 100 fs pulse with repetition of 82 MHz. The central energy of the laser pulse was varied in the energy region of the HH and LH excitons from 1.56 to 1.72 eV. The pump and probe beams were orthogonally polarized to each other, which results in elimination of the pump-beam contribution to the probe beam. The pump laser was focused onto the sample surface with the spot diameter of 300 μm. In the case of the pump power of 15 mW, which was a typical power in the present experiments, the pump density is 0.18 μJ/cm^2. Assuming that the excitonic absorption coefficient is about 1×10^4cm^{-1} (Ref. 13), the photoexcited carrier density is estimated to be 4.5×10^{15} cm^{-3}. Thus we can neglect Coulomb screening of excitons. The oscillation components were extracted by numerically differentiating time-resolved reflectivity changes in order to subtract a slowly varying back ground resulting from the relaxation of photoexcited carriers. In PLE measurements, the excitation light was produced by combination of a 100-W halogen lamp and a 32-cm single monochromator with a resolution of 0.5 nm. The emitted light was dispersed by a 25-cm double monochromator with a resolution of 0.5 nm and detected by a conventional photon-counting system with a cooled photomultiplier. The detection energy for PLE was slightly lower than the free exciton energy.

III. RESULTS AND DISCUSSION

Figure 1 shows the oscillatory profiles in the time derivatives of the time-resolved reflectivity changes in the (18,18) and (35,35) MQW's at 10 K. The energy of the laser pulse was tuned to the center energy between the HH and LH excitons in each sample. The oscillatory profiles observed in both the samples consist of two components. One is a strong oscillation that appears in the time range less than 1.5 ps, and the other is a weak oscillation observed over 4.0 ps. The periods of the strong oscillation are 109 fs and 275 fs for the (18,18) and (35,35) MQW's, respectively. In the time range longer than 1.5 ps, the period of the weak oscillation is 113 fs, which is the same in both the samples. The amplitude of the weak oscillation after 1.5 ps in the (18,18) MQW is about 20 times larger than that in the (35,35) MQW, while the amplitudes of the strong oscillations in both the samples are almost the same. In order to identify these oscillations and evaluate the intensities of the oscillation components, we divided the time-domain signals into two time ranges of 0.2–1.5 ps and 1.5–4.0 ps, and performed the Fourier transform (FT) of respective time-domain signals. Figures 2(a) and 2(c) show the FT spectra of the (18,18) and (35,35) MQW's in the time range from 0.2 to 1.5 ps, respectively. The peak frequencies of 9.2 and 3.6 THz correspond to ΔE_{HH-LH} in the (18,18) and (35,35) MQW's, respectively. Therefore, the oscillations observed in the time range less than 1.5 ps are attributed to the quantum beat of the HH and LH excitons. Figures 2(b) and 2(d) depict the FT spectra of the (18,18) and (35,35) MQW's in the time range from 1.5 to 4.0 ps, respectively. It is evident that peak frequencies are the same in both the samples: 8.8 THz corresponding to E_{LO} of GaAs. Thus, the long-lived oscillations in Fig. 1 arise from the coherent GaAs-like LO phonon. The ratio of the FT intensity of the coherent GaAs-like LO phonon to that of the excitonic quantum beat is 1.7×10^{-2} in the (18,18) MQW and 2.6×10^{-5} in the (35,35) MQW. The FT intensity, which corresponds to the square of the oscillation amplitude of the coherent GaAs-like LO phonon in the (18,18) MQW is about 650 times stronger than that in the (35,35) MQW.

In order to investigate the origin of the enhancement of the coherent GaAs-like LO phonon in the (18,18) MQW, the pump-energy dependence of the time-resolved reflectivity changes was observed in the (18,18) and (35,35) MQW's.

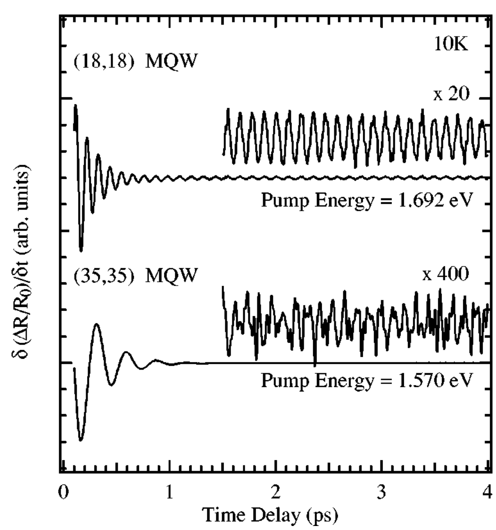

FIG. 1. Oscillatory profiles extracted by the time derivatives of the time-resolved reflectivity changes at 10 K, where the pump power was 15 mW. The upper and lower traces correspond to the reflectivity changes in the (18,18) and (35,35) MQW's, respectively. The pump energy corresponds to the center energy of the HH and LH excitons in each sample.

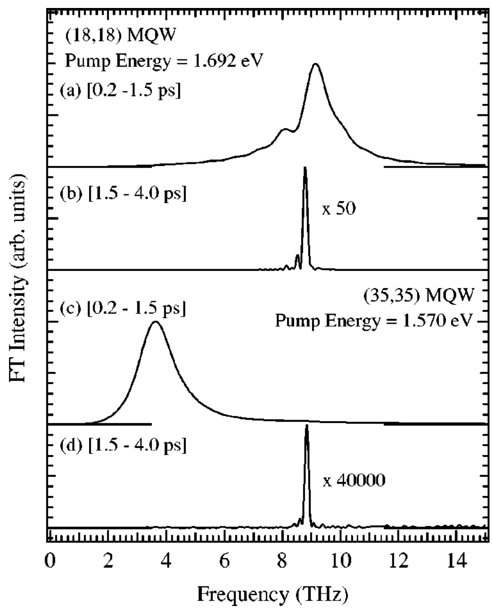

FIG. 2. FT spectra of the time-domain signals shown in Fig. 1 in two time ranges: (a) 0.2–1.5 ps in the (18,18) MQW, (b) 1.5–4.0 ps in the (18,18) MQW, (c) 0.2–1.5 ps in the (35,35) MQW, and (d) 1.5–4.0 ps in the (35,35) MQW.

The oscillatory profiles in the (18,18) MQW observed at various pump energies are shown in Fig. 3, where 1.671 and 1.708 eV correspond to the HH and LH exciton energies, respectively. It is evident that the amplitudes of the excitonic quantum beat and the coherent GaAs-like LO phonon depend on the pump energy. In order to clarify the pump-energy dependence of the intensities of the coherent oscillations, we performed the time-partition FT of the time-domain

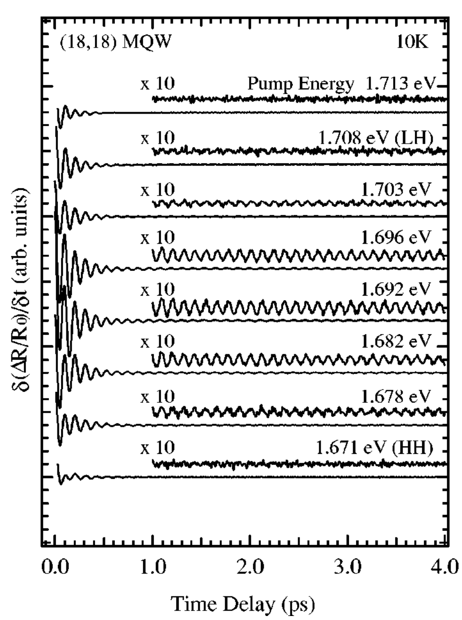

FIG. 3. Oscillatory profiles in the (18,18) MQW observed at various pump energies, where the pump power was 15 mW. The pump energies of 1.671 and 1.768 eV correspond to the HH and LH exciton energies, respectively.

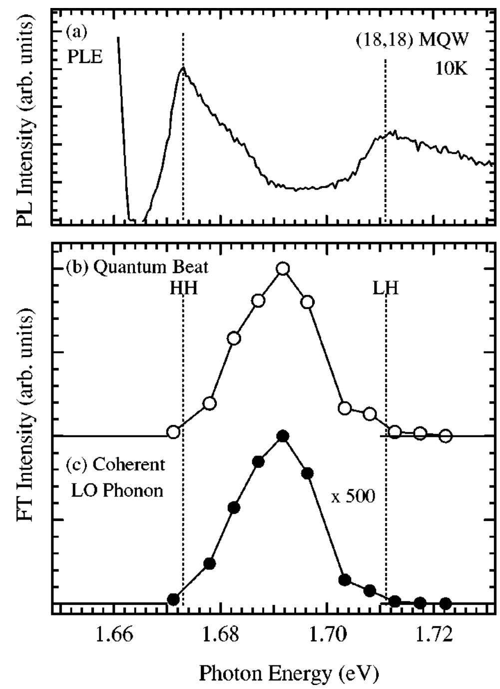

FIG. 4. (a) PLE spectrum in the (18,18) MQW, (b) integrated intensities of the FT bands of the excitonic quantum beat, and (c) that of the coherent GaAs-like LO phonon as a function of pump energy. The dotted lines indicate the HH and LH exciton energies.

signals in two time ranges: 0.2–1.5 ps for the excitonic quantum beat and 1.5–4.0 ps for the coherent GaAs-like LO phonon. In Figs. 4(b) and 4(c), the integrated intensities of the FT bands for the excitonic quantum beat and the coherent GaAs-like LO phonon are plotted as a function of pump energy, respectively. In addition, the PLE spectrum observed in the (18,18) MQW is shown in Fig. 4(a) for a reference, where the dotted lines indicate the HH and LH exciton energies. The integrated intensity of the excitonic quantum beat and that of the coherent GaAs-like LO phonon reach a peak at the same energy that is the center energy between the HH and LH excitons. Our finding is that the pump-energy dependence of the intensity of the coherent LO phonon is similar to that of the excitonic quantum beat in the case of $\Delta E_{HH-LH} \simeq E_{LO}$.

Figures 5(b) and 5(c) show the integrated intensities of the FT bands of the excitonic quantum beat and the coherent GaAs-like LO phonon as a function of pump energy, respectively, in the (35,35) MQW with $\Delta E_{HH-LH} \ll E_{LO}$. The PLE spectrum is shown in Fig. 5(a) for reference, where the dotted lines indicate the HH and LH exciton energies. The integrated intensity of the excitonic quantum beat reaches a peak around the center energy between the HH and LH excitons. On the other hand, the intensity of the coherent GaAs-like LO phonon shows the broad peak around the LH exciton energy. It should be noted that the pump-energy dependence of the coherent GaAs-like LO phonon in the (35,35) MQW is considerably different from that in the (18,18) MQW, which is a key point of the present work.

As mentioned above, the intensity of the excitonic quantum beat in both the MQW's exhibits the similar pump-

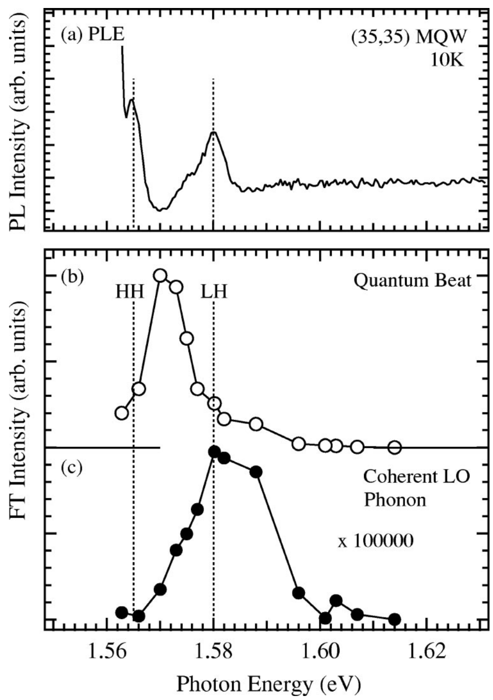

FIG. 5. (a) PLE spectrum in the (35,35) MQW. (b) integrated intensities of the FT bands of the excitonic quantum beat, and (c) that of the coherent GaAs-like LO phonon as a function of pump energy. The dotted lines indicate the HH and LH exciton energies.

energy dependence: the intensity reaches a peak around the center energy between the HH and LH excitons. This is a well-known feature of the excitonic quantum beat. On the other hand, with regard to the pump-energy dependence of the coherent GaAs-like LO phonon, the dependence of the (18,18) MQW shown in Fig. 4(c) is different from that of the (35,35) MQW shown in Fig. 5(c). If photoexcited carriers will take part in the generation of the coherent GaAs-like LO phonon, we can expect that the intensity is resonantly enhanced at the HH and LH exciton energies. The pump-energy dependence of the coherent GaAs-like LO phonon in the (35,35) MQW shows that the intensity is resonantly enhanced around the LH exciton energy. This result demonstrates the contribution of the photoexcited carriers to the generation process of the coherent GaAs-like LO phonon. However, there is no resonance effect around the HH exciton energy. The conclusive reason for the disappearance of the HH-exciton resonance is an open question at present. Unfortunately, there is no reference for the pump-energy dependence of the intensity of coherent LO phonons in quantum well systems. One of possible reasons may be attributed to the difference in the Bloch function forms of the HH and LH. The LH Bloch function has the growth-direction component, while the HH Bloch function has only the inplane component. The growth-direction component of the LH Bloch function may cause the resonance enhancement of the coherent GaAs-like LO phonon.

In the (18,18) MQW, the pump-energy dependence of the coherent GaAs-like LO phonon cannot be explained by the resonance effect described above, since the intensity reaches a peak around the center energy between the HH and LH excitons, which is similar to the pump-energy dependence of the excitonic quantum beat. This fact suggests the coupling between the excitonic quantum beat and the coherent GaAs-like LO phonon. Hereafter, we discuss the coupling mechanism. Under a condition that an internal electric field exists along the growth direction of the MQW, the longitudinal polarization is induced by the quantum beat of the HH and LH excitons, which leads to terahertz radiation.[9,10] The origin of the longitudinal polarization is attributed to the quantum-confined Stark effect[14] resulting in symmetry breaking of the HH and LH envelope functions: off-center configuration of the envelope function along the growth direction. In the present samples, there is no intentional electric field, but a surface electric field should exist because of pinning of the Fermi level at the surface, which is a usual nature of semiconductors. We observed photoreflectance (PR) signals of the HH and LH excitons (not shown here). It is well known that PR corresponds to noncontact electroreflectance utilizing modulation of a surface electric field by photoexcited carriers[15] and that PR signals of excitons are usually observed in quantum well system.[16] The electric field strengths in the present samples were estimated to be about 10 kV/cm from Franz-Keldysh oscillations of the band edge of GaAs substrate observed in PR signals.[17] This value is consistent with the results of GaAs/AlAs SL's.[18] Therefore, it is expected that the longitudinal polarization is caused by the excitonic quantum beat in the present samples. Since the coherent GaAs-like LO phonon in the MQW has also the longitudinal polarization along the growth direction, the excitonic quantum beat will couple with the coherent GaAs-like LO phonon via the longitudinal polarization under the condition that the energies of these coherent oscillations are very close to each other. Thus, it is considered that the pump-energy dependence of the intensity of the coherent GaAs-like LO phonon in the (18,18) MQW with $\Delta E_{HH-LH} \simeq E_{LO}$ is dragged by the excitonic quantum beat whose polarization is expected to be much larger than that of the coherent GaAs-like LO phonon from the oscillation intensities shown in Fig. 2. This leads to the enhancement of the coherent GaAs-like LO phonon by coupling with the excitonic quantum beat. The enhancement factor of the oscillation amplitude in the present study is about 25, which is larger than the factor of about 8 in the case of coupling of coherent LO phonons with Bloch oscillations.[5]

In order to confirm our consideration that the coherent GaAs-like LO phonon couples with the excitonic quantum beat, the pump-power dependence of the coherent GaAs-like LO phonon was measured in the (18,18) and (35,35) MQW's. The energy of the laser pulse was tuned to the center energy between the HH and LH excitons in each sample. Figure 6 shows the pump-power dependence of the FT intensity of the coherent GaAs-like LO phonon in the (35,35) and (18,18) MQW's. In the (35,35) MQW, the intensity of the coherent GaAs-like LO phonon increases in proportion to the square of the pump power. On the other hand, in the (18,18) MQW, the intensity of the coherent GaAs-like LO phonon linearly increases with increasing pump power. This difference in the pump-power dependence will reflect the difference in the generation mechanism of the coherent GaAs-like

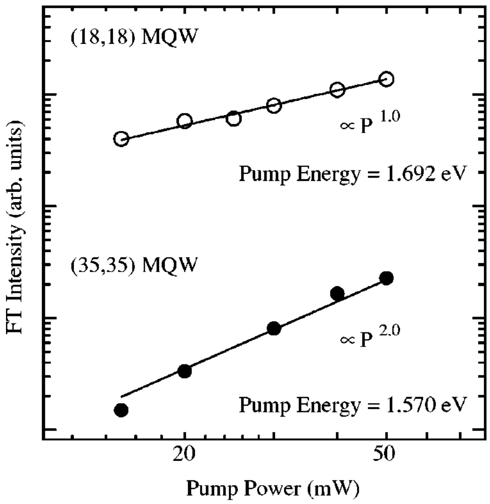

FIG. 6. FT intensity of the coherent GaAs-like LO phonon in the (18,18) and (35,35) MQW's as a function of pump power. The solid lines indicate the results of least-square fitting for the experimental data.

LO phonon. Takeuchi *et al.* investigated the coherent GaSb-like and AlSb-like LO phonons in GaSb/AlSb SL's.[12] They reported that the coherent GaSb-like (AlSb-like) LO phonon confined in the GaSb well layer (AlSb barrier layer) is generated through the DECP (ISRS) mechanism and that the FT intensities of both the GaSb-like and AlSb-like LO phonons are proportional to the square of the pump power. Yee *et al.* reported that the ISRS mechanism contributes the generation of the coherent GaAs-like LO phonon in GaAs/AlGaAs MQW's.[19] Since the pump-power dependence of FT intensity of the coherent GaAs-like LO phonon in the (35,35) MQW is square relation as shown in Fig. 6, the generation mechanism is considered to be the DECP and/or ISRS mechanisms. On the other hand, the pump-power dependence in the (18,18) MQW suggests that the generation of the coherent GaAs-like LO phonon is attributed to neither the DECP mechanism nor ISRS mechanism. Then, we consider the coupling between the excitonic quantum beat and the coherent GaAs-like LO phonon as the generation mechanism. As the exciton density that linearly depends on the pump power is increased, the amount of the longitudinal polarization due to the excitonic quantum beat is linearly increased. Assuming the linear coupling of the longitudinal polarization between the coherent GaAs-like LO phonon and the excitonic quantum beat, we can expect that the intensity of the coherent GaAs-like LO phonon is linearly increased with an increase in pump power. In fact, the FT intensity of the coherent GaAs-like LO phonon in the (18,18) MQW is proportional to the pump power as shown in Fig. 6. This result demonstrates that the longitudinal polarization of the excitonic quantum beat acts as a driving force for the coherent GaAs-like LO phonon under the coupling condition.

IV. CONCLUSIONS

We have investigated the coherent GaAs-like LO phonon and the quantum beat of the HH and LH excitons in the $(GaAs)_{18}/(AlAs)_{18}$ and $(GaAs)_{35}/(AlAs)_{35}$ MQW's by the reflection-type pump-probe technique. We find that the intensity of the coherent GaAs-like LO phonon in the (18,18) MQW with $\Delta E_{HH-LH} \simeq E_{LO}$ is markedly enhanced in comparison with that of the (35,35) MQW with $\Delta E_{HH-LH} \ll E_{LO}$. In order to reveal the mechanism of the enhancement of the coherent GaAs-like LO phonon, we have examined the pump-energy dependence and pump-power dependence of the coherent oscillations. In the (18,18) MQW, the pump-energy dependence of the intensity of the coherent GaAs-like LO phonon is similar to that of the excitonic quantum beat: the intensity reaches a peak at the center energy between the HH and LH excitons. On the other hand, in the (35,35) MQW, the pump-energy dependence of the coherent GaAs-like LO phonon shows a resonance profile around the LH exciton energy. The pump-energy dependence of the coherent GaAs-like LO phonon in the (18,18) MQW indicates that the coherent LO phonon couples with the excitonic quantum beat via the longitudinal polarization along the growth direction. It is revealed from the pump-power dependence of the FT intensity of the coherent GaAs-like LO phonon that the coherent LO phonon in the (35,35) MQW with $\Delta E_{HH-LH} \ll E_{LO}$ is generated through the well-known DECP and/or ISRS mechanisms leading to the square dependence. In the (18,18) MQW with $\Delta E_{HH-LH} \simeq E_{LO}$, the FT intensity of the coherent GaAs-like LO phonon is linearly increased with an increase in pump power, also suggesting the coupling between the excitonic quantum beat and the coherent LO phonon. It is concluded from the result described above that the excitonic quantum beat acts as a driving force for the coherent GaAs-like LO phonon under the condition of $\Delta E_{HH-LH} \simeq E_{LO}$, resulting in the enhancement of the coherent LO phonon.

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