Enhancement of coherent longitudinal optical phonon oscillations in a GaAs/AlAs multiple quantum well due to intersubband energy tuning under an electric field

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We have demonstrated the generation of intense coherent longitudinal optical (LO) phonon oscillations with use of intersubband energy tuning by application of electric field. The coherent LO phonon in a GaAs/AlAs multiple quantum well was investigated in detail as a function of electric field by using a reflection-type pump-probe technique. In the case that the intersubband energy is tuned to the LO phonon energy of GaAs by the electric field owing to a quantum confined Stark effect, the amplitude of the coherent LO phonon is intensively enhanced by a factor of 23 in comparison with the amplitude in a low electric field regime. This enhancement is caused by the coupling between the coherent LO phonon and the longitudinal polarization due to the impulsive interference between excitons with different subbands.

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Coherent phonon oscillations in nanostructured semiconductors,[1–4] e.g., multiple quantum wells (MQW's), superlattices (SL's), and quantum dots, have attracted much attention in the field of ultrafast phenomena from the viewpoint of dynamics of exciton-phonon and carrier-phonon interactions. Recently, the enhancement of coherent phonons in SL's and MQW's due to the coupling with coherent oscillations of photoexcited carriers and excitons, such as Bloch oscillations and quantum beats of the heavy-hole (HH) and light-hole (LH) excitons, has been reported as generation methods of intense coherent longitudinal optical (LO) phonons.[5–7] Dekorsy *et al.* reported on the coupling between the Bloch oscillation and the coherent 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] We reported the enhancement of the coherent LO phonon oscillations by tuning the frequency of the excitonic quantum beat to that of the coherent LO phonon.[7] This enhancement of coherent LO phonons results from the following two key factors: (i) The frequency of the excitonic quantum beat is almost equal to that of the LO phonon, a so-called resonant condition, and (ii) the excitonic quantum beat induces a longitudinal polarization oscillation along the growth direction of an MQW. Because of the resonant interaction between the excitonic quantum beat and coherent LO phonon, the excitonic quantum beat acts as a driving force for the coherent LO phonon.

The subband energies in an MQW can be controlled by an applied electric field, which is called a quantum confined Stark effect (QCSE),[8] as well as by a quantum size effect. Hence, it is possible to tune the intersubband energies to the LO phonon energy (E_{LO}). If two exciton states with different subbands are simultaneously excited by ultrashort pulses, it is expected that a quantum interference of the two exciton states occurs, which is well known as the quantum beat for the fundamental excitons with the quantum number $n=1$. In the case of the excitonic quantum interference associated with higher subbands with $n \geqslant 2$, it is assumed that a very fast dephasing prevents the quantum beat. That is the subject in the present work, which can be treated as an impulsive phenomenon. Under an applied electric field, the symmetry of the envelope functions of electrons and holes is broken by the QCSE, so that the excitonic quantum interference generates an impulsive longitudinal polarization along the growth direction of the MQW. Thus, we can expect the coupling between the excitonic quantum interference and the coherent LO phonon via the impulsive longitudinal polarization. In the present work, we have demonstrated the enhancement of the coherent GaAs-like LO phonons due to tuning the intersubband energy to the LO phonon energy with use of the QCSE in a GaAs/AlAs MQW. It has been found that the amplitude of the coherent GaAs-like LO phonon is enhanced intensively under the resonant condition that the energy spacing between the $n=1$ HH (HH1) and $n=2$ HH (HH2) subbands closely approaches to E_{LO}.

We used an undoped GaAs/AlAs MQW embedded in a *p*-*i*-*n* structure on a (001) n^+-GaAs substrate grown by molecular-beam epitaxy, where the *p* and *n* layers are 1 μm thick $Al_{0.5}Ga_{0.5}As$ layers with 50 nm undoped regions. The MQW consists of 20 periods of GaAs and AlAs layers whose thicknesses are 15.3 and 4.5 nm, respectively. The coherent LO phonon oscillation was 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 pump and probe beams were orthogonally polarized to each other in order to eliminate the pump-beam contribution to the probe beam. The typical pump energy was 1.571 eV, which is located between the transition energy from the HH1 subband to the $n=2$ electron (E2) subband and that from the HH2 one to E2 one in a high electric field regime. The pump density was kept at about 35 nJ/cm^2. Assuming that the excitonic absorption coefficient of the fundamental transition energy at zero electric field is about 1×10^4 cm^{-1} (Ref. 9), the photoexcited carrier density is estimated to be about 1×10^{15} cm^{-3}. Thus, formation of electron-hole plasma is negligible. We also performed photocurrent (PC) measurements at 10 K in order to estimate

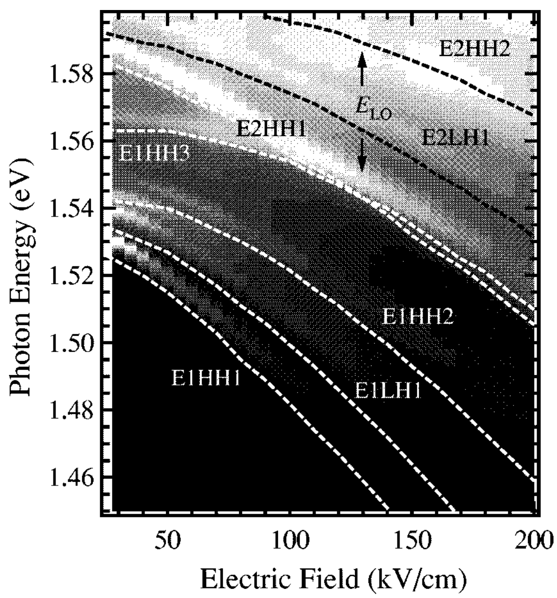

FIG. 1. Image map of the PC spectra of the GaAs(15.3 nm)/AlAs(4.5 nm) MQW at 10 K as a function of electric field, where the brightness is proportional to the PC intensity. The dashed curves show the transition energies calculated by using the TM method.

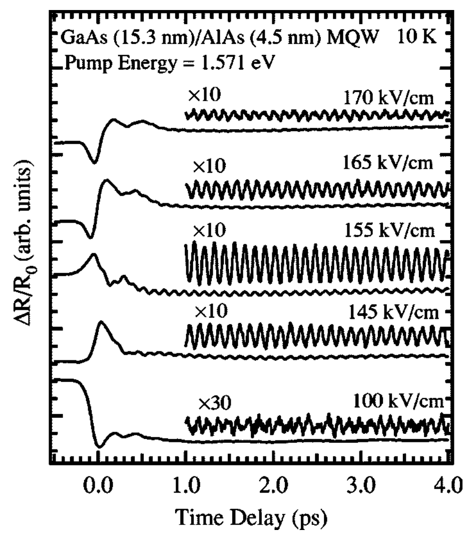

FIG. 2. Time-resolved reflectivity changes of the GaAs(15.3 nm)/AlAs(4.5 nm) MQW at various electric fields.

the energies of excitons including various higher subbands. The excitation light was produced by combination of a 100 W halogen lamp and a 32 cm single monochromator with a resolution of 0.3 nm.

Figure 1 shows the image map of PC spectra at various electric fields with a step of 5 kV/cm in the GaAs(15.3 nm)/AlAs(4.5 nm) MQW, where the brightness is proportional to the PC intensity. The value of the electric field F is estimated from $F=(V_b-V)/L$, where V_b is the built-in voltage of the p-n junction, V is the applied bias voltage, and L is the total length of the intrinsic layer: the MQW and undoped regions of the $Al_{0.5}Ga_{0.5}As$ layers. The dashed curves show the transition energies calculated by a transfer matrix (TM) method.[10] The values of the effective masses in the TM calculation are taken from Ref. 11. Many excitonic absorption peaks are observed. The origins of these PC peaks are assigned by comparison with the transition energies calculated by the TM method. EjHHi (EjLHi) indicates the transition from the HH (LH) subband with $n=i$ to the electron subband with $n=j$. The energy shifts of the PC peaks are due to the QCSE. The peak energies are consistent with the calculated results even in a high electric field regime. As the electric field is increased, the optical transitions between the electron and hole subbands with different quantum numbers, which are forbidden in principle at zero electric field, become obvious in the PC spectra, while the fundamental transitions of the E1HH1 and E1LH1 excitons are remarkably degraded. These changes of the transition probabilities originate from the symmetry breaking of the envelope functions of the electrons and holes due to the QCSE.

We can expect the coupling between the excitonic interference and coherent LO phonons via the longitudinal polarization under the condition that the energy difference between the two excitons with a common electron or hole subband approaches to the LO phonon energy. As shown in Fig. 1, since the energy difference between the E2HH1 and E2HH2 transitions at 130 kV/cm are almost equal to E_{LO} of GaAs, we can expect the coupling between the coherent GaAs-like LO phonon and the quantum interference of the E2HH1 and E2HH2 excitons. Thus, we tuned the pump energy to 1.571 eV, which is around the center energy between the E2HH1 and E2HH2 excitons at 130 kV/cm. It is noted that the fundamental excitons of E1HH1 and E1LH1, which are usually used for quantum-beat experiments, become unstable in such a high electric field regime as shown in Fig. 1, which is evident from the fact that the PC signals of the E1HH1 and E1LH1 excitons almost disappear.

Figure 2 shows the time-resolved reflectivity changes at various electric fields of the MQW sample. The highest electric field is 170 kV/cm in our measurement, because further increase of electric field induced instability of the results of pump-probe measurements. The time-domain signal observed at each electric field consists of the large reflectivity change around 0 ps and the long-lived oscillation. The initial part of the signal arises from a change of the carrier density. The shape of the initial part varies with an increase in electric field. This may be caused by the variation of the excess energy of the excitation relative to the E2HH1 exciton,[12] since the exciton energy is shifted by the QCSE. The oscillatory structure with the period of 113 fs in each signal lasts over 4.0 ps, and that is assigned to the coherent GaAs-like LO phonon. The amplitude of the coherent GaAs-like LO phonon changes with the electric field. We note that the amplitude of the coherent LO phonon is markedly enhanced around the electric field of 155 kV/cm. Since the quantum beat of the E2HH1 and E2HH2 excitons could not be observed, it seems that the dephasing times of the excitons are comparable to the excitation pulse width.

In order to estimate the intensity of the coherent GaAs-like LO phonon at each electric field, we performed the Fourier transform (FT) of the time-domain signals in the time range from 1.0 to 4.0 ps. Figure 3 shows the FT spectra at

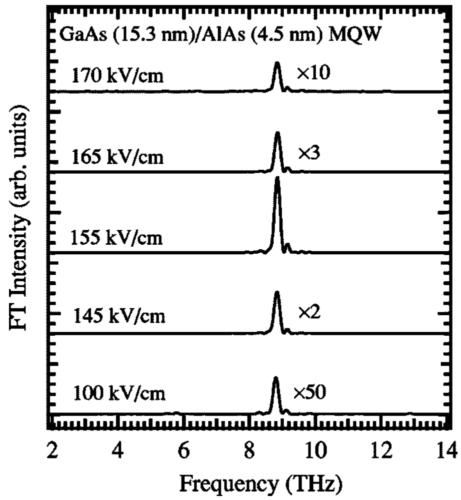

FIG. 3. Fourier-transform spectra of the time-domain signals shown in Fig. 2 in the time range from 1.0 to 4.0 ps at various electric fields.

various electric fields. It is evident that the peak frequency is constant at each electric field: 8.8 THz corresponding to E_{LO} of GaAs. It is obvious that the FT intensity is enhanced around 155 kV/cm. We plot the FT intensity of the coherent LO phonon and the energy difference between the E2HH1 and E2HH2 excitons ($\Delta E_{\mathrm{HH1-HH2}}$) estimated from the PC spectra as a function of electric field in Fig. 4, where the open and closed circles indicate the coherent LO phonon intensity and $\Delta E_{\mathrm{HH1-HH2}}$, respectively, and the broken line indicates E_{LO} of GaAs. The FT intensity, which corresponds to the square of the oscillation amplitude of the coherent LO phonon, reaches a peak at 155 kV/cm. The FT intensity of the coherent LO phonon at 155 kV/cm is about 500 times larger than that in a low electric field regime. The enhancement factor of the oscillation amplitude by tuning the HH1 - HH2 intersubband energy to E_{LO} is about 23, which is comparable to the enhancement factor of $\sim$25 by using the quantum beat of $n=1$ HH and LH excitons in a GaAs/AlAs MQW.[7]

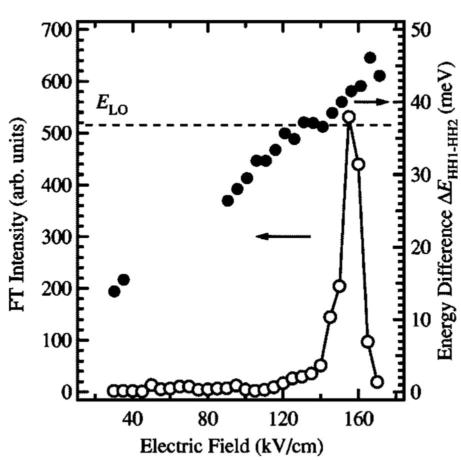

FIG. 4. Fourier-transform intensity of the coherent GaAs-like LO phonon and $\Delta E_{\mathrm{HH1-HH2}}$, which are indicated by open and closed circles, respectively, as a function of electric field. The dashed line indicates E_{LO} of GaAs.

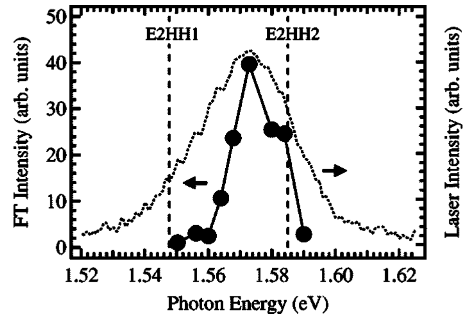

FIG. 5. Fourier-transform intensity of the coherent LO phonon at 155 kV/cm as a function of pump energy. The dashed lines indicate the energies of the E2HH1 and E2HH2 excitons obtained from the PC spectra. The dotted curve indicates the pump-laser spectrum with the center energy of 1.573 eV.

There are two possible reasons for causing the enhancement of the coherent LO phonons: (i) the resonance effect at the exciton energies and (ii) the coupling between the longitudinal polarization of the excitonic interference and that of the coherent LO phonon. In order to investigate the origin of the enhancement of the coherent LO phonon, the pump-energy dependence of the time-resolved reflectivity change was observed at 155 kV/cm that is the electric field for the largest enhancement. The pump-energy dependence of the intensity of the coherent LO phonon at 155 kV/cm is shown in Fig. 5. The dotted curve indicates the pump-laser spectrum with the center energy of 1.573 eV. If the excitonic resonance effect causes the enhancement of the coherent LO phonon, the pump-energy dependence should show three peaks at the E2HH1 (1.548 eV), E2LH1 (1.570 eV), and E2HH2 (1.585 eV) exciton energies that are obtained from the PC spectra. However, the resonance profile of the coherent LO phonon shows only one peak at 1.573 eV. This fact suggests that the coupling between the E2HH1-E2HH2 excitonic interference and the coherent LO phonon induces the enhancement of the coherent LO phonon. From an aspect of Raman scattering, the enhancement phenomenon of the coherent LO phonon is analogous to doubly resonant Raman scattering in a GaAs/AlGaAs quantum well,[13] where the energy spacing between the E1HH1 and E1LH1 excitons is equal to the GaAs-like LO phonon energy.

In Fig. 2, the oscillatory structure like the quantum beat corresponding to the excitonic interference between the E2HH1 and E2HH2 excitons is not observed. This disappearance is attributed to the short coherence time (ultrashort dephasing) of carriers at the higher subbands. Since the laser pulse has the spectral width broader than $\Delta E_{\mathrm{HH1-HH2}}$, these excitons are simultaneously generated by the laser pulse, and the impulsive longitudinal polarization will be generated through the interference between the E2HH1 and E2HH2 excitons. The impulsive longitudinal polarization due to the interference between the E2HH1 and E2HH2 excitons couples to the coherent LO phonon under the condition that $\Delta E_{\mathrm{HH1-HH2}}$ is almost equal to E_{LO} at 155 kV/cm, so that the resonant interaction between the coherent LO phonon and the longitudinal polarization of the excitonic interference enhances the coherent LO phonon. Although the electric field for the resonance between E_{LO} and the HH2-HH1 intersubband energy is estimated to be 130 kV/cm from the PC spectra as shown in Fig. 4, the electric field seems to be slightly shifted by Coulomb screening due to photoexcited carriers in the pump-probe measurement.

In summary, we have demonstrated that the coherent LO phonon in the GaAs/AlAs MQW is intensively enhanced by tuning the intersubband energy to the LO phonon energy with use of the QCSE. For the enhancement of the coherent LO phonon, the symmetry breaking of the envelope functions of electrons and holes due to the QCSE plays an important role producing the longitudinal polarization. Under the resonant condition between the intersubband energy, which is the HH1-HH2 energy spacing in the present work, and the LO phonon energy, the excitonic interference acts as a driving force to the enhancement of the coherent LO phonon. The present demonstration for the enhancement of the coherent LO phonon is applicable to various nanostructured semiconductors.

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