Coherent Oscillation of Zone-Folded Phonon Modes in GaAs-AlAs Superlattices

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We have observed a coherent oscillation of zone-folded acoustic phonons in GaAs-AlAs superlattices for the first time by means of a femtosecond time-resolved pump-probe technique. The oscillatory component in the time-resolved reflection signal corresponds to the upper branch (B_2 symmetry) of the first-order doublet mode. Carriers were selectively excited in well layers and the B_2-symmetry phonon mode was selectively generated. The dephasing time of the B_2 mode was measured to be 70 ps from our experiments.

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Progress in ultrafast laser techniques has enabled us to observe directly the coherent oscillation of phonon modes. Coherent phonons, that are impulsively generated by femtosecond pump pulses, modulate the dielectric constant and change the intensities of the transmitted and/or reflected light of the probe pulses. The ultrafast time-resolved measurement provides directly the amplitude, phase, and damping of the coherent phonons in various materials [1–7]. However, previous studies were mainly concentrated on bulk crystals, molecules, or amorphous materials. On the contrary, in semiconductor superlattices the Brillouin zone (BZ) is folded into a smaller mini-BZ, because of their long period along the growth direction. As a result of the zone-folding effect, new Raman modes appear and are called zone-folded modes [8,9]. In the present Letter, we report on a coherent oscillation of the zone-folded acoustic phonon mode in GaAs-AlAs superlattices which cannot be observed in bulk crystals.

Time-resolved pump-probe experiments were performed at room temperature with a mode-locked titanium-sapphire laser which produces 130 fs pulses at a repetition rate of 82 MHz in the tuning range of 720–800 nm. The pump and probe beams were orthogonally polarized to each other and were focused onto the sample with a spot size approximately 10 μm in diameter. The power of the pump and probe beams were 70 and 20 mW, respectively. The time delay of the pump beam was modulated by a shaker. The modulation amplitude and frequency of the shaker were 30 μm (100 fs) and 160 Hz, respectively. The probe beam passed through a variable optical delay line. The time derivative of the reflectivity change $d(\Delta R)/dt$ of the probe beam was measured by a lock-in amplifier with a p-i-n–Si photodiode. This time derivative detection technique is useful to observe the oscillations clearly [5,10]. Two samples of (15,15) with 80 periods, sample 1, and (18,18) with 60 periods, sample 2, grown on (001) GaAs substrates were used in this work, where the notation (m,n) indicates the numbers of monolayers of $(GaAs)_m$-$(AlAs)_n$ superlattices.

Figure 1 shows the time derivative traces of the reflectivity change for samples 1 and 2 at the laser wavelength 742 nm. In both samples an oscillatory structure is superimposed on a monotonically changing response. The insets show the traces near zero time delay and the signal is 2 orders of magnitude larger than the oscillation amplitude. Time integral of the traces, i.e., ΔR, deduced from the data rises within 200 fs and decreases with a decay time ~10 ps. This decay may be due to the cooling of the excited carrier distribution to the lattice temperature [11]. However, in this Letter we concentrate our attention to the oscillation component. The amplitude of

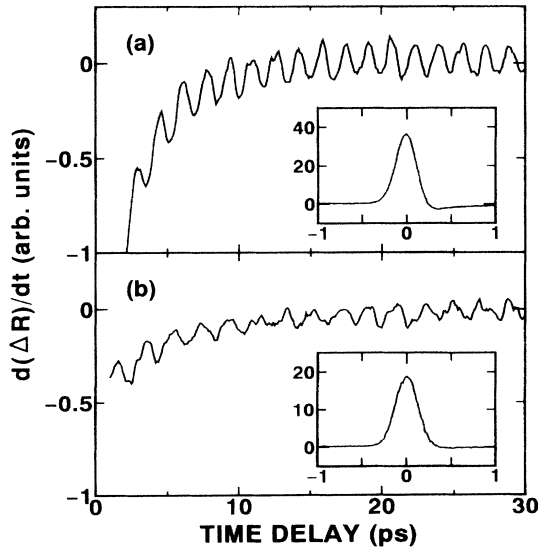

FIG. 1. Time derivative traces of the reflectivity change at a laser wavelength of 742 nm. (a) and (b) show the temporal traces for samples 1 and 2, respectively.

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the oscillation was on the order of $10^{-4}R_0$, where R_0 is the normal reflectivity. One can notice that the period of the oscillation for sample 1 is shorter than that for sample 2. This fact suggests that the oscillation originates from the periodicity of the superlattices and corresponds to the zone-folded acoustic phonon mode which was previously observed in Raman scattering experiments. The oscillatory component can be fitted by an expression for damped oscillations,

$$d(\Delta R)/dt = -A\exp(-t/\tau)\cos(\omega t + \varphi_0), \quad (1)$$

where ω is the phonon frequency, τ the phase relaxation time of the coherent phonon, and φ_0 a phase shift. Figure 2 shows the oscillatory component of sample 1 and the fitting curve shown by a solid line. We found that the time integral of the time derivative $d(\Delta R)/dt$, i.e., ΔR, has cosine behavior. In other words, ΔR has a peak at the zero time delay. The phase relaxation time was about 70 ps, which corresponds to the Raman linewidth of 0.15 cm^{-1}. This value is too narrow to be resolved by a conventional Raman spectrometer. The time-resolved technique enables us to observe the intrinsic dephasing time of the zone-folded mode. The dephasing time of the zone-folded mode is 1 order longer than the dephasing time of the LO phonon mode in bulk GaAs, that is a few picoseconds [2]. As will be shown later, the dispersion of the zone-folded modes suggests that there are few branches of phonon modes which have smaller energy than the created oscillation mode. Therefore, the zone-folded mode is hard to be scattered conserving the energy and momentum, and the dephasing time may be longer. Furthermore, over 100 ps of dephasing time was observed at 20 K.

Figure 3 shows the Fourier power spectra of the temporal traces shown in Fig. 1. One sharp peak is noticed in both the spectra. The peaks are located at (a) 20.5 cm^{-1} and (b) 16.9 cm^{-1}, which correspond to (a) 0.613 THz (1.63 ps) and (b) 0.507 THz (1.97 ps), respectively.

The dispersion curves of the zone-folded LA modes were calculated on the basis of the layered elastic continuum model by Rytov [12]. The dispersion is written by

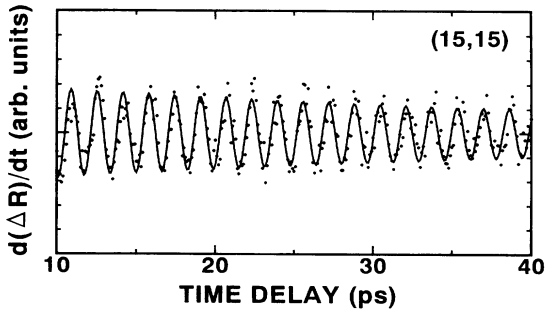

FIG. 2. The oscillatory component of the time derivative trace of the reflectivity change in sample 1. The solid line is a fit to the experimental data described in the text.

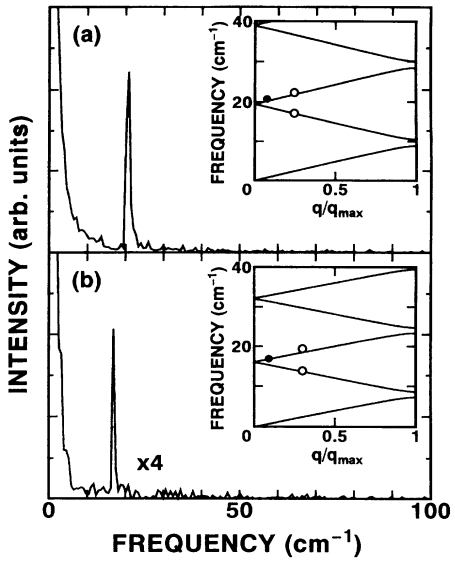

FIG. 3. Fourier power spectra of the temporal traces shown in Fig. 1. The insets show phonon dispersions of the two samples based on the Rytov model, where q_{max} is the zone-edge wave vector π/d and d is the period of the superlattices. The open circles of the insets indicate the first-order doublet peaks obtained through Raman scattering experiments at an excitation wavelength 514.5 nm and the solid circles indicate the peak frequencies of the power spectra.

$$\cos(qd) = \cos\left(\frac{\omega d_1}{\nu_1}\right)\cos\left(\frac{\omega d_2}{\nu_2}\right) - \frac{1+\kappa^2}{2\kappa}\sin\left(\frac{\omega d_1}{\nu_1}\right)\sin\left(\frac{\omega d_2}{\nu_2}\right), \quad (2)$$

where ω and q are phonon frequency and the superlattice wave vector, ν_1 and ν_2 are the sound velocities of GaAs and AlAs, d_1 and d_2 the thicknesses of the two constituting layers, and d the period of the superlattices defined by $d = d_1 + d_2$. The coefficient κ is defined by $\kappa = \nu_1\rho_1/\nu_2\rho_2$, where ρ_1 and ρ_2 are the GaAs and AlAs densities, respectively. The insets of Fig. 2 show phonon dispersion curves of the two samples calculated by using Eq. (2), where q_{max} is the zone-edge wave vector π/d. We used the values $\nu_1 = 4.726 \times 10^5$ cm/s and $\rho_1 = 5.3149$ g/cm^3 for GaAs, and $\nu_2 = 5.118 \times 10^5$ cm/s and $\rho_2 = 3.7285$ g/cm^3 for AlAs [13]. The open circles in the insets indicate the first-order doublet peaks obtained by Raman scattering experiments at an excitation wavelength 514.5 nm and the solid circles indicate the peak frequencies of the power spectra. The observed data agree well with the calculated curves. The Raman spectrum of the first-order zone-folded mode shows doublet peaks, whereas the coherent oscillation has one component, which corresponds to the upper branch of the doublet mode.

The lower and upper branches of the doublet mode at $q = 0$ have A_1 and B_2 symmetries, respectively [13]. In both samples, the thickness d_1 and d_2 are almost the

same. Hence, the lattice oscillation of the first-order A_1 mode has a node at the center of the constituting layers, whereas that of the first-order B_2 mode has a node at the interfaces of the layers. At our experimental wavelength, the femtosecond laser pulses excite the carriers in the well layers (GaAs layers) selectively. The optically excited carriers probably induce a stress in the well layers and the B_2-symmetry phonon mode was selectively generated. It is noted that when the thickness of the constituting layers d_1 and d_2 are not the same, the node of the B_2-symmetry mode is not located at the interface. In this case, it is expected that the A_1-symmetry mode will be excited more effectively and the beat of the A_1- and B_2-symmetry modes will be observed.

Furthermore, we measured the coherent oscillation by varying the laser wavelength from 720 to 800 nm. Figure 4 shows the temporal traces of sample 1 for various laser photon energies. Each of the broken lines indicates a local minimum point of the oscillation in the trace of the laser photon energy 1.67 eV and it is noted that the phase of the oscillation shifts by π around the energy of 1.6 eV. Using Eq. (1), we estimated the amplitude of the oscillation as a fitting parameter. Figure 5(c) shows the estimated amplitude as a function of the laser photon energy, where the change of the sign indicates the π phase shift. The lowest $1s$ heavy-hole exciton energy is 1.62 eV and the absolute value of the amplitude enhances around the absorption peak. This fact shows that substantial absorption is necessary to excite the B_2-symmetry phonon mode.

The π phase shift was also obtained in sample 2. We found that there was no relationship between the energy position of the π phase shift and the exciton energy. It is known that the reflectivity spectra of the superlattices are

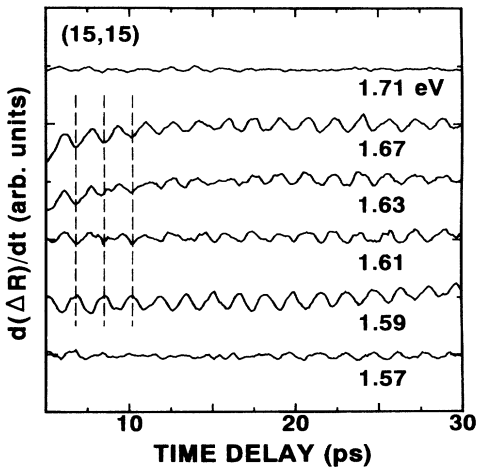

FIG. 4. Time derivative traces of the reflectivity change in sample 1 for the various laser photon energy. Each of the broken lines indicates a local minimum point of the oscillation in the trace of the laser photon energy 1.67 eV. The phase of the oscillation shifts by π around the photon energy 1.6 eV.

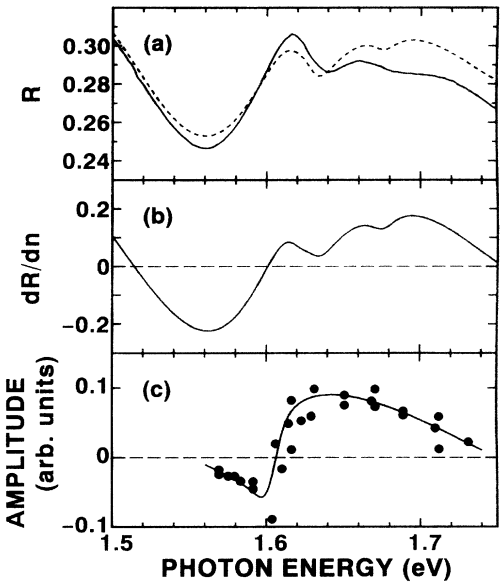

FIG. 5. (a) Experimental (solid line) and theoretical (broken line) reflectivity spectra in sample 1. (b) Theoretical calculation of dR/dn as a function of the photon energy. (c) Amplitude of the oscillation as a function of the laser photon energy. The change of the sign indicates the π phase shift. The solid line is a guide to the eye.

seriously modified by multiple reflections of light, if the total thickness of the superlattice layer is on the order of light wavelength [14]. Thus we take into account multiple reflections to explain the π phase shift. Figure 5(a) shows the reflectivity spectrum of sample 1. The broken line shows the theoretical calculation taking into account multiple reflections and it explains well the characteristics of the experimental spectrum. Here the absolute value of the experimental reflectivity is adjusted because of the experimental uncertainty. The refractive index n is considered to be modulated by the coherent phonon and then the reflectivity is also modulated. The reflectivity change ΔR can be described as $\Delta R = (dR/dn)\Delta n$, where Δn is the change of the refractive index. If the sign of Δn changes, this probably occurs around the exciton energy, because n itself changes drastically around it. However, the π phase shift did not correlate with the exciton energy. Thus, the possibility of the sign change of Δn is excluded and the sign change of dR/dn may explain the observed π phase shift. We calculated numerically the sign of dR/dn considering the multiple reflection effect. Figure 5(b) shows the energy dependence of dR/dn and two dips are due to the heavy- and light-hole excitons. It is noted that the sign of dR/dn changes around 1.6 eV. This change of the sign is considered to be the origin of the π phase shift.

In conclusion, we have reported on the coherent oscillation of zone-folded phonon modes in GaAs-AlAs superlattices by means of a femtosecond time-resolved pump-probe technique in the reflection configuration. One oscillatory component was dominantly observed in

the time derivative traces of the reflectivity change. The oscillatory component corresponds to the upper branch (B_2 symmetry) of the first-order zone-folded doublet mode. These results suggest that the femtosecond laser pulses excite the well layers selectively and induce coherent phonons with B_2 symmetry. The intrinsic dephasing time was estimated in our experiments and was about 70 ps at room temperature. Furthermore, when the laser photon energy was changed, π phase shift of the oscillation was observed. This phase shift could be explained by multiple reflection in a superlattice thin film. In semiconductor superlattices, many confined optical phonons as well as zone-folded acoustic phonons are observed in Raman scattering experiments. When we use a shorter pulse laser, it may be possible to observe the confined optical phonon oscillations. So far, many kinds of superlattice systems have been fabricated. Our work opens the way to study directly the dynamical phonon properties and interaction between coherent phonons and optically excited carriers in these semiconductor superlattice systems.

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