Observation of coherent folded acoustic phonons propagating in a GaAs/AlAs superlattice by two-color pump-probe spectroscopy

K. Mizoguchi,* M. Hase, and S. Nakashima
Department of Applied Physics, Osaka University, Yamadaoka 2-1, Suita 565-0871, Japan

M. Nakayama
Department of Applied Physics, Osaka City University, Sugimoto, Sumiyoshi-ku, Osaka 558-8585, Japan

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The coherent folded longitudinal acoustic (FLA) phonons in a GaAs/AlAs superlattice have been studied using a two-color pump-probe technique and a designed sample in which the superlattice layer is sandwiched by $Al_xGa_{1-x}As$ cladding layers. The two-color pump-probe technique makes it possible to separate regions of generation and detection of coherent phonons, which provides a powerful tool to examine the spatial dynamics of coherent phonons. We have found two types of coherent FLA phonons: the first-order FLA phonons with finite wave vectors ($q \neq 0$) and the zone-center mode of the first-order FLA phonons ($q = 0$). When ballistic acoustic phonons are generated near the surface region of the $Al_xGa_{1-x}As$ cladding layer and propagate through the superlattice layer, the first-order FLA phonons with $q \neq 0$ are observed in the superlattice layer. On the other hand, when the FLA phonons are generated within the superlattice layer, the zone-center FLA phonon extending from the superlattice layer is detected near the surface region of the $Al_xGa_{1-x}As$ cladding layer. [S0163-1829(99)15135-X]

I. INTRODUCTION

A pump-probe technique with an ultrashort pulse laser is a useful tool to study the dynamics of carriers and phonons.[1–3] The propagation of the longitudinal acoustic (LA) phonons generated in materials has been studied by using a one-color (1C) pump-probe technique.[4–11] The coherent folded LA (FLA) phonons with a finite wave vector ($q \neq 0$) in superlattices (SLs) have also been observed by the 1C pump-probe technique.[5,7–11] Recently, acoustic phonons emitted from a single quantum well which propagate to the sample surface have been measured by using a two-color (2C) pump-probe technique with the near-IR pump and violet probe pulses;[12] however, the propagation of coherent FLA phonons has not been studied by the 2C pump-probe measurement. In order to understand the space evolution of the coherent FLA phonons with $q \neq 0$, it is necessary to examine the propagation of the coherent phonons by separating the generation and detection regions of the coherent phonons. This separation is realized by using the 2C pump-probe technique and a SL sample with a designed structure. In the 1C pump-probe technique, the regions of the detection and generation of the phonons are the same because the wavelengths of the pump and probe light are identical.

In the present work, we have focused on the propagation of coherent FLA phonons in a GaAs/AlAs SL sandwiched by $Al_xGa_{1-x}As$ cladding layers. The near-IR light of a Ti:sapphire pulse laser and the second-harmonic light (violet light) of the fundamental laser pulse are used for 2C measurements. The sample consists of a GaAs/AlAs SL and $Al_xGa_{1-x}As$ cladding layers. The SL layer used is opaque for the 2C beams, while the $Al_xGa_{1-x}As$ cladding layer is transparent for the near-IR light and opaque for the violet light. Thus, the generation region of the coherent phonons can be selected by the wavelength of the pump pulse in the 2C pump-probe experiment. For example, when using the violet pump and the near-IR probe, the coherent phonons are generated near the surface and are detected in the whole SL layer by the near-IR probe light. We have found two different types of the FLA modes for different generation regions. The phonon related to the zone-center mode appears for the near-IR pump and violet probe configuration, while the mode with $q \neq 0$ appears for the violet pump and near-IR probe configuration. In addition, we have observed phonons in the lowest LA branch, which corresponds to the unfolded LA branch, for both of the pump excitation energies.

II. EXPERIMENTAL PROCEDURE

The sample used was a $(GaAs)_{24}(AlAs)_8$ SL sandwiched between $Al_{0.4}Ga_{0.6}As$ cladding layers grown on a (001) GaAs substrate, where the subscript denotes the number of the GaAs or AlAs monolayers (thickness of one monolayer is equal to 0.283 nm). The total thickness of the SL layer which consists of 40 periods of GaAs/AlAs layers is about 362 nm and the thickness of the $Al_xGa_{1-x}As$ cladding layers is 500 nm. The sample structure is schematically depicted in the inset of Fig. 1. The two interfaces between the SL and $Al_xGa_{1-x}As$ cladding layers exist at $d = 500$ and 862 nm, where d is the depth from the surface of the $Al_xGa_{1-x}As$ cladding layer. Hereafter, we will call the $(GaAs)_{24}(AlAs)_8$ SL ‘‘(24, 8) SL.’’

The reflection-type pump-probe experiments were performed at liquid N_2 temperature by using a mode-locked Ti:sapphire laser delivering about 70-fs pulses. In the 2C pump-probe measurement, one of two beams was the fundamental laser light with the wavelength of 777 nm tuned to the first interband transition of the SL, which was determined by a photoreflectance experiment. The other was the second-

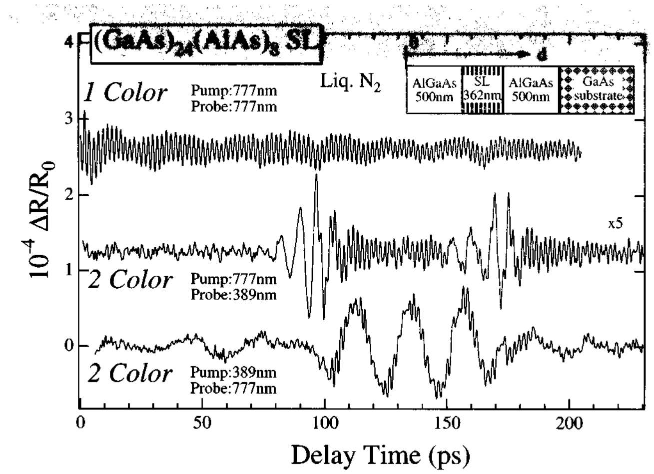

FIG. 1. The oscillatory parts of the time-resolved reflectivity changes of the (24,8) SL observed by the 1C and 2C pump-probe measurements. Inset is the sample structure prepared in a GaAs/AlAs superlattice.

harmonic light of the fundamental laser light (389 nm) produced by a frequency doubling crystal (BBO) with a thickness of 0.5 mm. In addition, the 1C pump-probe measurement was performed with the fundamental laser light in order to obtain the standard information of the coherent phonon. The pump (probe) powers were adjusted to 70 mW (5 mW) in both cases of the 777-nm-pump/389-nm-probe and 389-nm-pump/777-nm-probe experiments. Hereafter, we will call the 777-nm-pump/389-nm-probe and 389-nm-pump/777-nm-probe ''2C(777, 389)'' and ''2C(389, 777),'' respectively. The probe beam was delayed by a variable optical delay line in a range of 0–240 ps. The oscillation components were numerically extracted by subtracting a slowly varying background resulting from the relaxation of photoexcited carriers.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 1 shows the time-resolved oscillatory reflectivity changes for the (24,8) SL obtained by using the 1C and 2C pump-probe techniques. The band-gap energy of the $Al_{0.4}Ga_{0.6}As$ cladding layer is about 2.01 eV at 77 K,[13] being larger than the energy of the fundamental laser light. The penetration depth of the 389-nm light estimated from the absorption coefficient ($\sim 5\times 10^5$ cm^{-1}) of $Al_{0.4}Ga_{0.6}As$ is about 20 nm.[14] For the 2C(389,777) experiment, the phonons are generated near the surface of the $Al_xGa_{1-x}As$ cladding layer, and the signals are probed in the SL layer, and vice versa for the 2C(777,389) experiment. For the 1C time-domain spectrum, the oscillatory structure with a weak beat is observed. The period of the main oscillation is about 1.9 ps and the relaxation time exceeds 150 ps.[8] It is noted that the strong reflectivity change due to the large refractive-index discontinuity at the surface was observed at the delay time around 0 ps. In Fig. 1, the signals were excluded because it is not related to the coherent phonons.

For the 2C(777,389) time-domain spectrum, the oscillations with the short period of about 1.9 ps are observed in the time range after about 95 ps. Moreover, the oscillating transients with the period of about 7 ps appear around 95 and 170 ps. The time of 95 (170) ps corresponds to the passing time of the LA phonons propagating in the region of $d=0$–500 (0–862) nm, because under the above pump-probe condition the coherent phonons are generated in the SL layers and are detected near the surface of the $Al_xGa_{1-x}As$ cladding layer. The passing times calculated using sound velocities of $v_s=5.03\times 10^5$ cm/s for the SL layer and 4.90×10^5 cm/s for the $Al_xGa_{1-x}As$ cladding layer are 99 and 173 ps for $d=0$–500 and 0–862 nm, respectively. In this calculation, the SL layer is regarded as an $Al_{0.25}Ga_{0.75}As$ mixed-crystal layer and the sound velocities of the mixed crystals are estimated by linear interpolation from those of GaAs and AlAs.[13] The passing time of the LA phonons is consistent with that reported by Baumberg *et al.*, who have measured the propagation of the LA phonon pulse emitted from a quantum well.[12]

On the other hand, in the 2C(389,777) signal, the weak oscillations with a period of about 25 ps are observed in the time regions before about 90 ps and after about 180 ps (Fig. 1). A strong oscillation with a period of about 25 ps, which accompanies the weak oscillations with the higher frequencies, is observed only in the time range from about 90 to 180 ps. The amplitude of the weak oscillations with a 25 ps period seems to be enhanced in the time range from 90 to 180 ps. The week oscillations in the time region before 90 ps and after 180 ps are due to the interference between light beams reflected by the moving phonon pulse (strain pulse) at different depths, the sample surface, and the GaAs substrate.[4,6] The enhanced oscillations in the time range from 90 to 180 ps arises from the LA phonons propagating in the SL layer, which are generated near the surface of the $Al_xGa_{1-x}As$ cladding layer. Such enhancement of the reflectivity change observed is similar to the resonance of the oscillation amplitude of the coherent FLA phonons excited at the energy gap of the SL layer.[8,11]

We have precisely determined the frequencies of the coherent phonons observed in the SL from the Fourier transform (FT) of the time-domain data as depicted in Fig. 2. The 1C-FT spectrum shows that a strong sharp peak (0.53 THz) accompanies two weak peaks at the lower- and higher-frequency sides (0.50 and 0.60 THz). The strong peak is located at a frequency slightly lower than the central position of the weak satellite peaks. In the 2C(777,389)-FT spectrum, the sharp peak at 0.53 THz appears dominantly but the satellite peaks do not. The calculation of the propagation times described above indicates that the 0.14 THz peak results from the propagating LA phonons generated around the head and tail of the interfaces between the SL and $Al_xGa_{1-x}As$ cladding layers ($d=500$ and 862 nm). Moreover, the fine peaks which are located at 0.013-THz intervals are observed around 0.14 THz. This fine structure arises from the FT of the time intervals of two phonon pulses at 95 and 170 ps. In the 2C(389,777)-FT spectrum, the sharp peak at 0.53 THz is not observed and only two satellite peaks appear. The frequencies of the satellite peaks (0.50 and 0.60 THz) are the same as those in the 1C-FT spectrum. The strong peak at 0.04 THz arises from the LA phonon oscillation during the propagation through the SL layer (90–180 ps).

The dispersion curves of the zone-folded LA modes were calculated on the basis of an elastic continuum model.[15] Figure 3 shows the phonon-dispersion curves of the (24,8) SL,

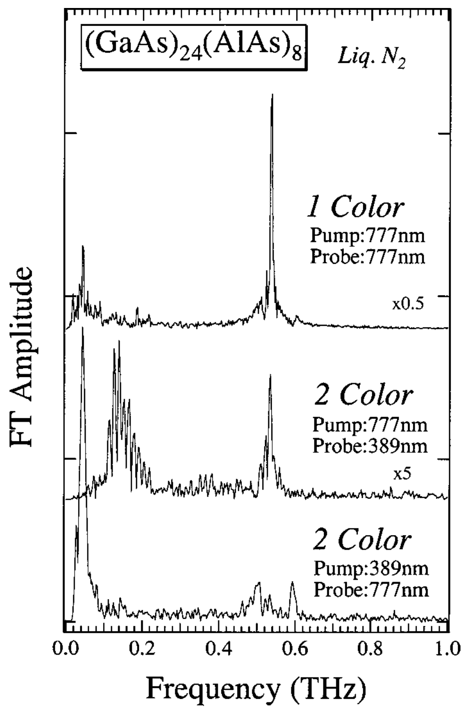

FIG. 2. Fourier transform spectra of temporal traces in the (24,8) SL observed by the 1C and 2C techniques.

where q_{max} is the zone-edge wave vector π/D and D is the period of the SL. The closed circles, the open triangles, and the open squares indicate the peak frequencies observed in the 1C-, 2C(777,389)-, and 2C(389,777)-FT spectra, respectively. The vertical lines in Fig. 3 show the wave vectors given by $q/q_{max}=4nD/\lambda$, where n is the refractive index at

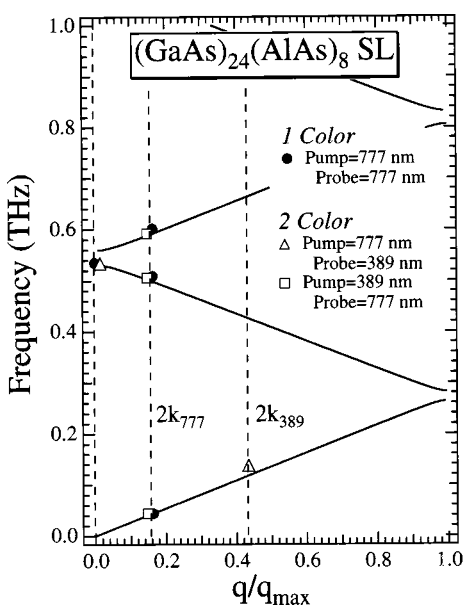

FIG. 3. Phonon dispersions of the (24,8) SL based on the elastic continuum model. The closed circles indicate the peak frequencies of the FLA and unfolded LA phonons observed in the 1C-FT spectrum. The open triangles and open squares indicate the peak frequencies observed in the 2C(777,389)- and the 2C(389,777)-FT spectra, respectively.

the wavelength λ of the laser pulse (389 and 777 nm). We assume that the refractive index of the SL corresponds to that of $Al_{0.25}Ga_{0.75}As$, which is estimated by linear interpolation for those of $Al_{0.198}Ga_{0.802}As$ and $Al_{0.315}Ga_{0.685}As$:[14] $n=3.5$ and 4.4 for 777 and 389 nm, respectively. From a comparison between the peak frequencies and the calculated dispersion curves of the FLA modes, the peaks with the frequencies of 0.53, 0.50, and 0.60 THz observed in the 1C and 2C experiments are attributed to the first-order FLA modes with $q=0$ and $q\neq0$. The peaks with 0.04 and 0.14 THz originate from the unfolded LA mode with $q\neq0$. Moreover, it is found that the wave vector of the FLA and unfolded LA modes with $q\neq0$ depends on that of the probe pulses in the 2C pump-probe experiment: $q=2k_{probe}$. Similar results have been observed in a $(GaAs)_{24}(AlAs)_{24}$ SL having no $Al_xGa_{1-x}As$ cladding layers.[16]

The group theory analysis of the FLA phonon indicates that the lower and upper branches of the first-order FLA mode at the zone center ($q=0$) have A_1 and B_2 symmetries, respectively: The order of these symmetries is reversed for the second-order FLA modes. Near the zone center the lower and upper branches have the symmetry of mixed A_1 and B_2. In the 1C- and 2C-FT spectra, the coherent FLA phonons with B_2 symmetry at the zone center are not observed, but those with A_1 symmetry which are Raman active are observed.[17]

If the coherent phonon is generated by the pump pulses via the impulsive stimulated Raman scattering process,[3,10] the wave vector of the coherent FLA phonons with $q\neq0$ generated in the SL layer should be determined by the pump beam. The 2C(389,777)-FT spectrum indicates that the wave vector of the coherent phonons observed by the 2C pump-probe technique is related to the detection process of the coherent phonon and that the wave vector of the FLA phonons depends on that of the probe beam. In the 2C(389,777) experiment, the pump pulse produces a high density of electrons and holes near the surface region because the penetration depth of 389 nm is about 20 nm for the $Al_xGa_{1-x}As$ cladding layer. We consider that the instantaneous surface-potential bending due to the photoexcited carriers near the surface region[18,19] will generate a phonon pulse with wave vectors distributing in the q space. The generated phonon pulse propagates through the $Al_xGa_{1-x}As$ cladding layer onto the SL layer. When the phonon pulse arrives at the SL layer, it modulates the dielectric constants of the SL via its strain field: The elastic strain would produce the LA modes peculiar to the SL layer. The phonon pulse which has various wave vectors might include a component with an energy of 0.6 THz at least for generation of the coherent FLA phonons with 0.6 THz. The FLA phonons induced by the phonon pulse are the propagating phonons with various wave vectors. The coherent FLA phonon with a certain wave vector would be detected by phase-matching with the probe pulses. One of the possible explanations for the detection process of the FLA phonon with $q\neq0$ is the stimulated Raman process of the probe pulse or phase-dependent Raman emission.[20] This was experimentally suggested by the observation that the coherent phonon spectra in heme proteins of biomolecules, light-harvesting pigments, and dye molecules depend on the energy of the probe light.[21–25]

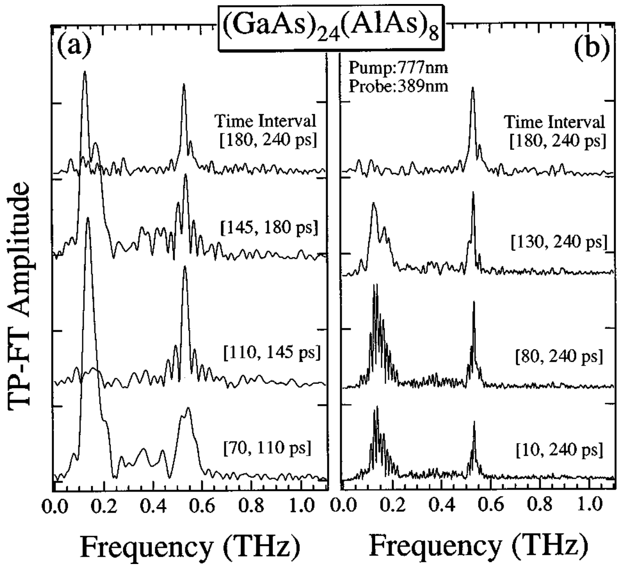

FIG. 4. Time-partitioning FT spectra of the time-domain signal with the 2C(777,389) experiment for the different time intervals of (a) $[t_0 - t_1]$ and (b) $[t_2$–240 ps].

In the 2C(777,389) configuration, where the coherent phonon is probed around the surface of the $Al_xGa_{1-x}As$ cladding layer, the 0.53-THz mode corresponding to the FLA mode at $q=0$ in the SL layer is observed. It is usually recognized that the mode at $q=0$ is the standing wave. We have studied the dynamic process of the phonon with 0.53 THz, which is observed after about 95 ps as shown in Fig. 1, by means of the time-partitioning FT (TP-FT) of the coherent oscillations for different time intervals.[26] Figures 4(a) and 4(b) show the TP-FT spectra in the time range of $[t_0 - t_1]$ and $[t_2$–240 ps], respectively, where the time range in Fig. 4(a) is sequential and no peaks in the TP-FT spectrum for [10–70 ps] were observed. The strong peaks at around 0.14 THz observed only in the time ranges of [70–110] and [145–180 ps] are due to the phonon pulses emitted from the interfaces between the SL and $Al_xGa_{1-x}As$ cladding layers ($d=500$ and 862 nm). The peak at 0.53 THz is observed in the early time range of [70–110 ps]. This result indicates that the phonon with the frequency of 0.53 THz is produced at a time that is the same as the generation time of the 0.14-THz phonon pulse, and that the two phonon modes propagate together to the surface. As shown in Fig. 4(b), after 180 ps, the broad peak around 0.14 THz disappears and only the peak at 0.53 THz is observed. This result indicates that the phonon with 0.53 THz remains even after the phonon pulse emitted from the tail of the SL layer ($d=862$ nm) arrives at the surface (180 ps), and that the phonon mode corresponding to the $q=0$ FLA mode in the SL layer propagates through the $Al_xGa_{1-x}As$ cladding layer onto the surface. The propagation of the $q=0$ FLA mode, which corresponds to the standing wave in the SL, may be explained as follows. The symmetry of the observed $q=0$ FLA mode is A_1 as described above. The amplitude of the A_1 mode does not have a node at the interface between the SL and $Al_xGa_{1-x}As$ cladding layers,[17] so that the vibration of the $q=0$ FLA mode is transferred to that of the bulk LA mode with the same frequency in the $Al_xGa_{1-x}As$ cladding layer. Then, the bulk LA phonon induced by the $q=0$ FLA mode propagates through the $Al_xGa_{1-x}As$ cladding layer.

As discussed above, the wave vector of the coherent phonon is determined by that of the probe light: $q=2k_{\text{probe}}$. From this scenario, it is expected that in the 2C(777,389) reflection-type experiment, the $q\neq 0$ is observed and the $q=0$ mode is not detected; however, the experimental result of the first-order FLA mode in the 2C(777,389) configuration is contrary to this expectation. The amplitudes of the FLA modes with $q\neq 0$ may be too small to be observed in our experimental setup, because the 777-nm-pump light excites entirely the SL region and the standing wave is dominantly generated in the SL layer. The observation of the $q=0$ mode suggests that the detection mechanism of the $q=0$ modes is different from that of the $q\neq 0$ mode. For the $q\neq 0$ mode, the propagating wave packet generated by the pump light consists of whitelike FLA phonons with various wave vectors, so that the wave-vector selection of $q=2k_{\text{probe}}$ is required for the detection. On the other hand, the $q=0$ mode generated in the SL layer is monochromatic at the surface and will directly modulate the probe light. One of the possible mechanisms for the modulation is photoelastic effects.[4–6,11] The relative amplitudes of the FLA modes with $q=0$ and $q\neq 0$ observed by the 1C pump-probe technique are consistent with the Raman intensities calculated using the photoelastic model.[11] In the 2C pump-probe experiment, the generation and detection areas are separated, so that it is difficult to discuss the amplitude of the FLA modes with $q=0$ and $q\neq 0$ at present.

IV. SUMMARY

The use of the 2C pump-probe technique and the SL sample with a designed structure enables us to select the generation and detection regions of the FLA phonons propagating in the SL sample. We have observed the two different types of the coherent FLA phonons by switching the generation and detection regions of the coherent phonon: The propagating phonon related to the coherent FLA phonon mode with $q=0$ is observed near the surface for the 2C(777,389) configuration, while the FLA modes with $q\neq 0$ in the SL layer are detected for the 2C(389,777) configuration. It follows from the observation that the strain field by the phonon pulse which is generated near the interface between the SL and $Al_xGa_{1-x}As$ cladding layers produces coherent phonons. The 2C pump-probe experiments demonstrate that the wave vector of the coherent phonon with $q\neq 0$ is determined by that of the probe pulses ($q=2k_{\text{probe}}$).

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*Present address: Department of Applied Physics, Osaka City University, 3-3-138 Sugimoto, Sumiyoshi-ku, Osaka 558-8585, Japan.

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