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Dynamical Characteristics of a Coherent Longitudinal Optical Phonon in a GaAs Buffer Layer Optically Covered with a GaSb Top Epitaxial Layer Investigated with Use of Terahertz Spectroscopy

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Abstract. We demonstrate that, in a GaSb/GaAs epitaxial structure, a coherent longitudinal optical (LO) phonon in a GaAs buffer layer optically covered with a GaSb top layer is observed utilizing terahertz spectroscopy. It is confirmed from Raman scattering measurements that only the optical phonons in the GaSb layer is optically observable. In the terahertz-wave measurement, the Fourier power spectrum of a terahertz waveform exhibits both the coherent GaAs and GaSb LO phonon bands; namely, the coherent LO phonon in the optically covered GaAs buffer layer is observed in the terahertz-wave measurement. This fact demonstrates that the instantaneous surface potential modulation, which originates from the impulsive carrier excitation by the pump beam, reaches the GaAs buffer layer. Consequently, the above-mentioned surface potential modulation generates the coherent GaAs LO phonon that cannot be optically excited. In addition, we perform a time-partitioning Fourier transform analysis in order to investigate the decay dynamics of the coherent GaAs and GaSb LO phonons. The decay times of the coherent GaAs and GaSb LO phonons are estimated to be 2.0 and 3.3 ps, respectively. The longer decay time of the coherent GaSb LO phonon is attributed to the fact that the phonon density of state in a final state of the decay process of GaSb is relatively small in comparison with that of GaAs.

Keywords: Terahertz spectroscopy; Coherent longitudinal optical phonon; Phonon decay dynamics; GaAs; GaSb
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INTRODUCTION

Terahertz spectroscopy, which is a kind of time-domain femtosecond spectroscopy, is an meaningful tool for investigation of characteristics of coherent phonons.[1, 2] In emission processes of terahertz electromagnetic waves from longitudinal optical (LO) phonons, initially, illumination of a femtosecond pulse pump beam generates carriers. Following the illumination, the surge current of the photogenerated carriers instantaneously modulates a surface band-bending potential through a screening effect. Finally, the instantaneous band-bending modulation induces the coherent LO phonons, which emit the terahertz waves.[3] The above-mentioned terahertz-wave emission mechanism suggests that the substantial driving force of the coherent LO phonon is not the pump-beam illumination but the subsequent band-bending modulation caused by the surge current. Accordingly, it leads to an expectation that, even in an under layer optically covered with upper layers, the terahertz wave from the coherent LO phonons is generated through a change in the band bending. This expectation, however, has not been examined in the earlier works.[4-7] We also, here, emphasize that terahertz spectroscopy features a time-domain measurement. The feature of terahertz spectroscopy provides us information on dynamical characteristics, *e.g.*, the decay time, of the coherent LO phonons.

In the present work, initially, we explored the feasibility of detecting the terahertz wave from the coherent LO phonon in an undoped GaAs buffer layer optically covered with a GaSb top layer in a GaSb/GaAs epitaxial structure. We succeeded in detecting the terahertz wave from the coherent LO phonon in the GaAs buffer layer in addition to the coherent GaSb LO phonon in the top layer. The simultaneous detection of both the coherent LO phonons indicates the appropriateness of the above-mentioned expectation. Next, we estimated the decay times of the coherent GaAs using a time-partitioning Fourier transform analysis. We found that the decay time of the coherent GaSb LO phonon is longer than that of the coherent GaAs LO phonon. We discuss the difference in the decay times between the coherent GaAs and GaSb LO phonons from the viewpoint of a phonon density of state (DOS) related to the decay process of the coherent LO phonons.

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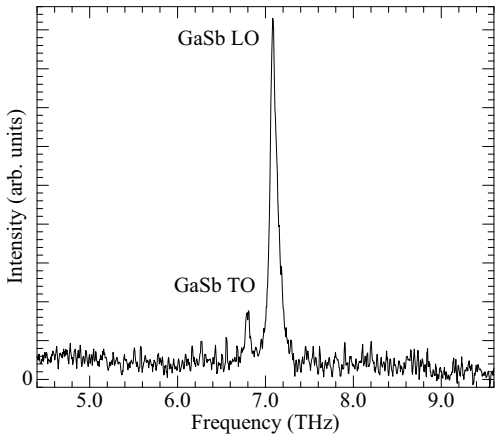

FIGURE 1. Raman scattering spectrum of the GaSb/GaAs structure measured at room temperature.

EXPERIMENTAL PROCEDURE

The present sample, the GaSb/GaAs epitaxial structure, was grown on a (001)-oriented semi-insulating GaAs substrate by molecular beam epitaxy. The thicknesses of the GaSb top layer and GaAs buffer layer were 900 nm and 200 nm, respectively. The both layers were undoped. Consequently, the free carrier absorption of terahertz waves is negligible in the GaSb top layer, which means that the GaSb layer is transparent to terahertz waves.

In advance to a terahertz wave measurement, we performed a Raman scattering measurement of the GaSb/GaAs structure in the back scattering configuration at room temperature in order to confirm that the GaAs layer is optically covered by the GaSb layer. The photon energy and power of the excitation beam were 1.58 eV and 19 mW, respectively.

The time-domain terahertz-wave measurement was performed at room temperature. Humidity was kept at about 10% during the measurement by purging with dry nitrogen gas. The pump beam was focused on the sample with the incidence angle of 45°. The diameter of the beam spot on the sample surface was about 100 μm. The emitted terahertz wave was collected with use of two off-axis parabolic mirrors, and was detected by an optically gated dipole antenna with a gap of 6.0 μm formed on a low-temperature-grown GaAs layer. The duration time of the laser pulses was about 50 fs. The powers of the pump and gate beams were fixed to 120 mW and 10 mW, respectively. The repetition of the laser pulses was 90 MHz. The photon energies of both the beams were the same value of 1.57 eV. This value is almost the same as the excitation photon energy in the Raman scattering measurement.

EXPERIMENTAL RESULTS AND DISCUSSION

Simultaneous Detection of the Coherent GaAs and GaSb LO Phonons

Figure 1 shows the Raman scattering spectrum of the present sample. Two bands are observed at the frequencies of 6.8 and 7.1 THz. The frequencies of the GaSb transverse optical (TO) and LO phonons are 6.8 and 7.0 THz at the Γ point of the Brillouin zone, respectively.[8] Accordingly, the observed Raman bands are assigned to the GaSb TO and LO phonons in order of frequency. The GaAs LO phonon at the Γ point with a frequency of 8.8 THz (Ref. 9) is not observed in the Raman scattering spectrum. The present result is reasonable because the penetration length of the excitation beam is estimated to be 160 nm in the GaSb layer with use of the absorption coefficient [10]; namely, the GaSb top layer optically covers the GaAs buffer layer. We also note that, in principle, the TO-phonon band is forbidden in ideally (001)-oriented zinc-blende crystals. The observation of the GaSb TO-phonon band indicates the presence of an offset orientation along the [111] or [110] direction in the GaSb layer because, in zinc-blend crystals, the appearance of the Raman scattering band of a TO phonon is allowed in the (111) and (110) faces. The

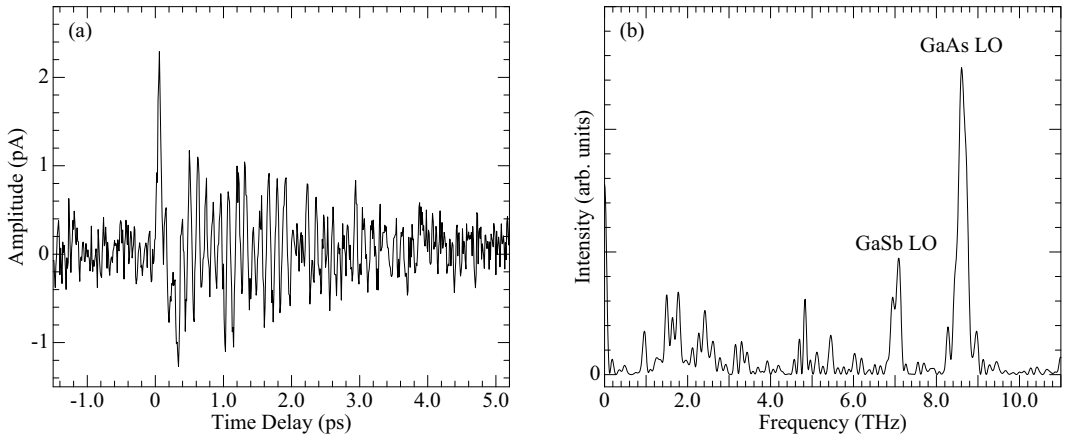

FIGURE 2. (a) Amplitude of the terahertz waveform of the GaSb/GaAs structure as a function of time delay at room temperature. (b) Fourier power spectrum of the terahertz waveform in Fig. 2(a).

responsible factor is attributed to the lattice mismatch between GaAs (lattice constant: 0.565 nm) and GaSb (0.610 nm) [ref. 8] that causes the above-mentioned the offset orientation. It is, therefore, considered that a certain amount of misorientation induced by the lattice mismatch exists in the GaSb/GaAs interface.

The terahertz waveform of the GaSb/GaAs structure is shown in Fig. 2(a). The monocycle signal, which results from the surge current of photogenerated carriers,[11] appears around the time delay of 0 ps. The monocycle signal is followed by an oscillatory profile. It is evident that the oscillatory profile shows a beat pattern. The appearance of the beat indicates that multiple oscillation modes are observed.

In order to analyze the observed modes, we applied the Fourier transform to the terahertz waveform. The Fourier power spectrum is shown in Fig. 2(b). The band corresponding to the monocycle signal weakly appears around 2.0 THz. The band at 7.1 THz is assigned to the coherent GaSb LO phonon. The noteworthy phenomenon is the appearance of the GaAs LO phonon at 8.6 THz. Taking account of the fact that the frequency of the GaAs LO phonon in a single crystal is 8.8 THz, the shift of the phonon frequency $\Delta\omega$ is -0.2 THz in the present GaAs buffer layer. The value of $\Delta\omega$ corresponds to the tensile strain of 1.8×10^{-2}, assuming the biaxial strain induced by the lattice mismatch. In the estimation of the strain, the phonon deformation potential was taken from Ref. 12. We, therefore, conclude that the terahertz wave from the coherent GaAs LO phonons is observable even in the GaAs buffer layer optically covered with the GaSb layer.

Next, we discuss the generation mechanism of the coherent GaAs LO phonon. In general, compound semiconductors have a surface potential-bending potential resulting from the surface Fermi level pinning.[13] It is considered that, in an undoped semiconductor crystal, the region of the surface potential bending extends to several micrometers in the internal side of the crystal because carriers originating from unintentional dopants hardly screen the surface potential bending. The present GaSb/GaAs structure has a total thickness of 1.1 μm; consequently, the surface potential bending reaches the GaAs buffer layer. The illumination of the pump beam generates dense carriers around the surface region, and, as a result, causes the instantaneous surface potential modulation across the GaSb/GaAs interface. This phenomenon is a responsible factor of generating the coherent GaAs LO phonon.

Decay Process of the Coherent GaAs and GaSb LO Phonons

Next, we estimate the decay times of the coherent GaAs and GaSb LO phonons, using a time-partitioning Fourier transform analysis.[14] The time-partitioning Fourier power spectrum $I(\omega)$, where ω is a frequency, is given by the following equation:

$$I(\omega) = \left| \int_{\tau}^{8\,\mathrm{ps}} A(t) \exp(-i\omega t) dt \right|^2 \qquad (1)$$

Here, $A(t)$ is the terahertz waveform and τ is the time delay (-2 ps ≤ τ < 8 ps) determining the time window of the time-partitioning Fourier transform.

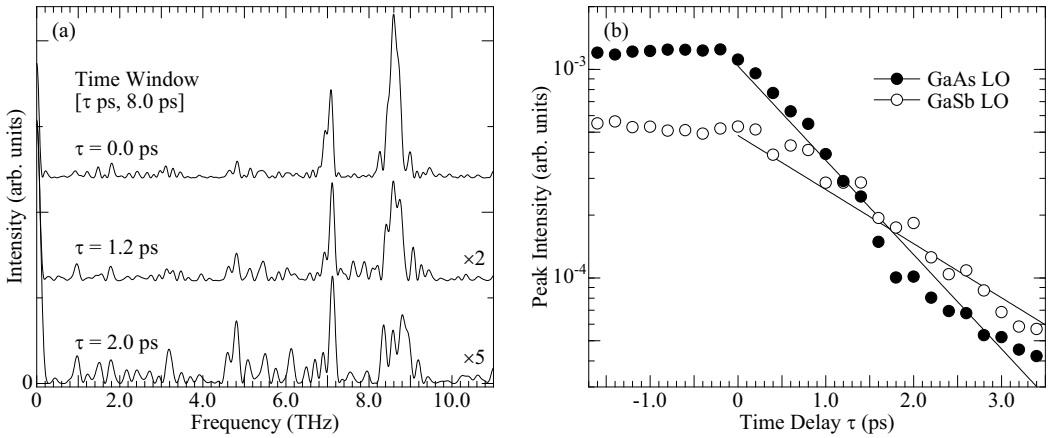

FIGURE 3. (a) Time-partitioning Fourier power spectra using Eq.(1). The values of τ = 0.0, 1.2 and 2.0 ps (b) Peak intensities of the GaAs and GaSb LO phonon bands plotted as a function of time delay τ. The closed and open circles denote the plots of the peak intensities of the coherent GaAs and GaSb LO phonon band, respectively. The solid lines are the fitted results using a single exponential function.

The time-partitioning Fourier power spectrum is shown in Fig. 3(a). As shown in Fig. 3(a), the peak intensity of the coherent GaAs LO phonon band rapidly decreases in comparison with that of the coherent GaSb LO phonon band. At τ = 2.0 ps, the peak intensity of the coherent GaSb LO phonon exceeds that of the coherent GaAs LO phonon. In order to estimate the decay rate of the coherent LO phonon band, we plotted the peak intensities of the coherent GaAs and GaSb LO phonon bands as a function of τ, which are shown in Fig. 3(b). The peak intensities of the LO phonon bands start decreasing at 0 ps. We performed fitting to the date using a single exponential function to evaluate the decay rate. The fitted results are depicted as the solid lines. The estimated decay rates are 1.0 and 0.60 ps^{-1} for the coherent GaAs and GaSb LO phonons, respectively. Note that, as indicated in Eq. (1), the decay rate is estimated from the Fourier power spectrum that corresponds to the square of the amplitude. Consequently, the decay rate of the amplitude of the terahertz wave from the coherent LO phonon, which is shown in Fig. 2(a), is a half value of the decay rate estimated from the time-partitioning Fourier transform method. Thus, the decay rates of the terahertz waves from the coherent GaAs and GaSb LO phonons are about 0.50 and 0.30 ps^{-1}, respectively, which correspond to the decay times of 2.0 and 3.3 ps, respectively.

Here, we discuss a difference in the decay times between the GaAs and GaSb LO phonons. The decay time connects with the anharmonic decay process of the LO phonon. According to Ref. 15, interactions between the LO phonon and other two phonons, which are governed by the third-order anharmonicity, are the most probable process contributing to the phonon-decay time. The phonon-decay mechanism strongly depends on each DOS of final two phonons; therefore, the dominant contributions are expected to result from the relaxation channels involving phonons at the minimal or maximal points of dispersion curves in momentum space. This is because the phonon DOS is relatively large at the minimal or maximal points. In GaAs, the LO phonon at the Γ point, the LO(Γ) phonon, decays into transverse acoustic (TA) and LO phonons at the L point,[15] where the frequencies of the TA and LO phonons are about 60 cm^{-1} (1.8 THz) and 235 cm^{-1} (7.1 THz) at the L point,[15] respectively. The sum of the frequencies of the TA(L) and LO(L) phonons is 8.9 THz, which almost agrees with the frequency of the LO(Γ) phonon: 8.8 THz. Accordingly, in the decay process into the LO(L) and TA(L) phonons, the energy and momentum conservation rules are kept. Here, we verify whether the above-mentioned relaxation channel, LO(Γ) into LO(L) and TA(L), involves the minimal or maximal points of the dispersion curves in momentum space. Figure 4 shows the calculated phonon dispersion relation in GaAs.[16] As shown in Figure 4, the GaAs LO(L) and TA(L) phonons locate at the minimal and maximal point of the dispersion curves, respectively. Accordingly, the decay channel of GaAs LO(Γ) phonon into GaAs LO(L) and TA(L) is relatively wide.

There is no report on the relaxation channel of the GaSb LO(Γ) phonon. In order to discuss the relaxation channel, the calculated phonon dispersion relation, which is taken from Ref. 17, is shown in Fig. 5. According to Fig. 5, the frequencies of GaSb LO(L) and TA(L) are 6.1 and 1.5 THz, respectively. The sum of the frequencies of the TA(L) and LO(L) phonons 7.6 THz: The L point locates at [0.5, 0.5, 0.5]. The value of the sum is much larger than the frequency of the GaSb(Γ) phonon: 7.0 THz. Consequently, in GaSb, the relaxation into the LO(L) and TA(L) phonons is forbidden from the viewpoint of the energy conservation rule. This means that the decay channel of the

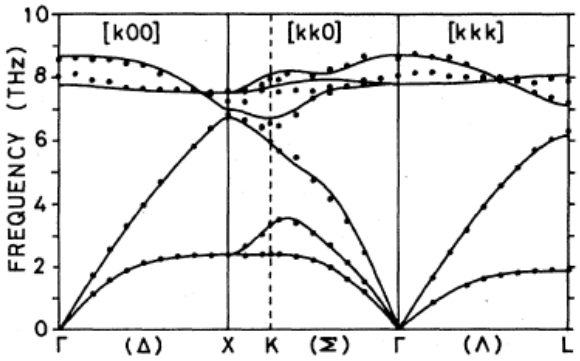

FIGURE 4. Phonon dispersion curve in GaAs. The symbols are the experimental data obtained with use of neutron scattering, while the solid curve is the fitted result. Reprinted figure with permission from Ref. 16. Copyright (1985) by the American Physical Society.

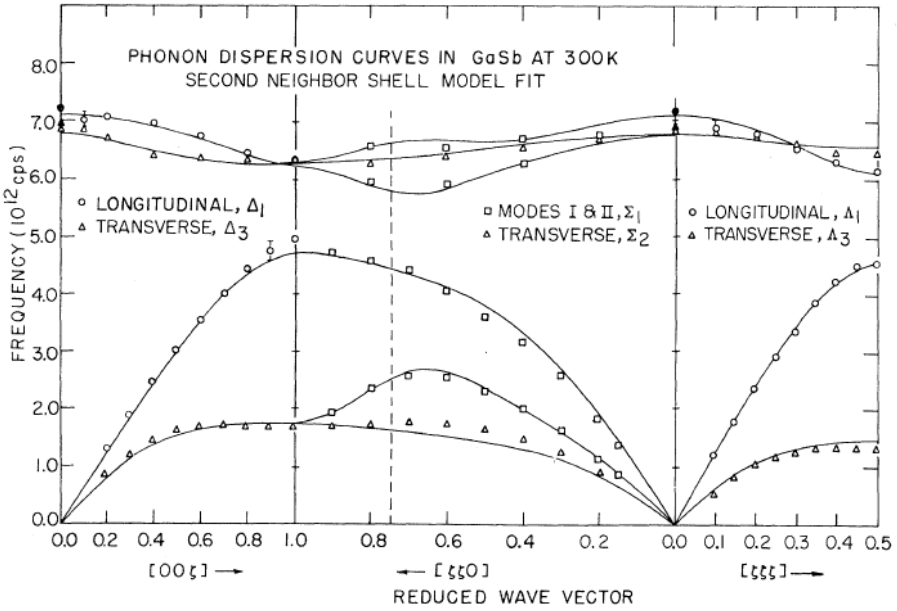

FIGURE 5. Phonon dispersion curve in GaSb. The symbols are the experimental data obtained with use of neutron scattering, while the solid curve is the fitted result. Reprinted figure with permission from Ref. 17. Copyright (1975) by the American Physical Society.

GaSb(Γ) phonon is different from that of the GaAs(Γ) phonon. Here, we focus our attention on the longitudinal-and-transverse-mixing acoustic phonon diversion curve along the [ζ, ζ, 0] direction. The frequencies of the IA and IIA phonons at $\zeta = 0.7$ are 2.6 and 4.4 THz, respectively, where the IA and IIA phonons are the lower and upper branch modes, respectively. The sum of these values is the same as the frequency of the LO(Γ) phonon: 7.0 THz. This means that the decay channel from the LO(Γ) photon to the IA and IIA phonons keeps the momentum conservation rule; namely, the IA and IIA phonons are the most suitable for the decay of the LO(Γ) photon. As shown in Fig. 5, the IIA phonon dispersion curve has a maximal point at $\zeta = 0.7$. In contrast, the IIA phonon dispersion curve does not have a maximal nor minimal points at $\zeta = 0.7$; namely, the DOS of the IIA phonon is relatively small at $\zeta = 0.7$. It is, therefore, expected that the decay time of the GaSb LO(Γ) phonon is longer than that of the GaAs LO(Γ) phonon. The above consideration coincides with the experimental result that the decay time of the GaSb LO(Γ) phonon (3.3 ps) is 1.6 times longer than that of the GaAs LO(Γ) phonon (2.0 ps). The definite assignment of the

relaxation channel requires the temperature-dependence measurement of the phonon-decay time,[15] though we could not perform this measurement.

SUMMARY

We have investigated the Raman scattering spectrum and terahertz wave from the GaSb/GaAs epitaxial structure. In the Raman scattering spectrum, the GaSb LO-phonon band is observed, whereas the GaAs LO-phonon band is not detected. This is because the penetration length of the excitation beam is too short to generate the phonons in the GaAs layer. In contrast, the Fourier power spectrum of the terahertz wave exhibits both the coherent GaAs and GaSb LO phonons. The responsible factor for the generation of the coherent GaAs LO phonons has been attributed to the phenomenon that the instantaneous surface potential modulation, which originates from the impulsive carrier excitation by the pump pulses, reaches the GaAs buffer layer. From the above-mentioned comparison, we conclude that the terahertz-wave measurement of the coherent LO phonon is applicable to the investigation of the dynamical phenomena in the layers optically covered with the upper layer. We have also estimated the decay times of the coherent GaAs and GaSb LO phonons with use of the time-petitioning Fourier transform analysis. The decay times of the coherent GaAs and GaSb LO phonons are estimated to be 2.0 and 3.3 ps, respectively. The longer decay time of the coherent GaSb LO phonon has attributed to the relatively small phonon DOS connecting with the dispersion curve.

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