Observation of coherent acoustic phonons in Fibonacci superlattices

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Coherent acoustic phonons are observed in the femtosecond time-resolved reflectivity of GaAs/AlAs Fibonacci superlattices. The time-domain data reveal a complicated superposition of many folded acoustic modes induced by the quasiperiodicity of the Fibonacci sequence. We discuss the phonon spectra from the viewpoint of the polarizability modulation due to the acoustic phonons on the basis of a photoelastic model. In addition, we demonstrate that the resonance of the heavy-hole exciton transition remarkably modifies the oscillation amplitude of the coherent phonons. [S0163-1829(97)00615-2]

The study of quasiperiodic structures realized in semiconductor heterostructures has been a subject of great interest since the quasiperiodicity introduces self-similarity and fractal behavior in the electronic and vibrational properties of these systems.[1] The effects of the quasiperiodicity on acoustic phonons in Fibonacci superlattices have been extensively investigated by Raman spectroscopy.[2–7] It has been revealed that the quasiperiodicity causes a series of doublet Raman bands of acoustic phonons that are quite similar to folded modes induced by the formation of the miniature Brillouin zone in periodic superlattices.[3–7] Although the comprehension of Raman spectra of acoustic modes in quasiperiodic superlattices has been established, there has been no report on time-domain data directly related to the understanding of phonon dynamics.

The advance of a pump-probe technique using ultrashort light pulses has enabled us to study the dynamics of coherently excited phonons in bulk GaAs,[8] GaAs/AlAs periodic superlattices,[9] and $GaAs/Al_xGa_{1-x}As$ heterostructures.[10] Here we report on the observation of coherent acoustic phonon modes in quasiperiodic GaAs/AlAs Fibonacci superlattices. An application of a high-sensitivity data-acquisition system enables us to trace the coherent dynamics over more than 100 ps, thus obtaining unprecedented high-resolution phonon spectra in the subtetrahertz frequency range. The complicated structure of the time-domain data exhibits the quasiperiodic nature in the frequency domain. We analyze the profiles of the Fourier spectra on the basis of a photoelastic model,[11] which has been successfully applied to explain the Raman profiles of acoustic phonons in Fibonacci superlattices.[3–7] Moreover, it is found that the amplitude of the coherent oscillations is remarkably affected by the resonance of the heavy-hole exciton transition.

The Fibonacci superlattices under investigation are quasiperiodic heterostructures composed of alternating the quantum wells and/or barriers in accordance with the Fibonacci sequence

$$S_{N+1}=\{S_N,S_{N-1}\} \quad \text{with} \quad S_1=\{A\}, \quad S_2=\{B\},$$

where S_N is the Nth finite realization of the lattice. The quasiperiodicity of the Fibonacci sequence provides δ-function peaks in reciprocal space that have self-similar features. Two different Fibonacci superlattices grown on a (001) GaAs substrate were investigated. The first sample consists of $A=(GaAs)_{10}/(AlAs)_{10}$ and $B=(GaAs)_{10}$ and the second sample is defined by $A=(GaAs)_8/(AlAs)_5$ and $B=(GaAs)_8$, where the subscript indicates the layer thickness in the units of monolayer (one monolayer is equal to 0.283 nm). Both superlattices consist of the 13th order of the Fibonacci sequence. During the molecular-beam epitaxy (MBE) growth the intensity oscillations of reflection high-energy electron-diffraction patterns were used to control the layer thickness. The period of the intensity oscillation exactly corresponds to the time of monolayer growth; therefore, the fluctuation of layer thicknesses in the samples is expected to be less than one monolayer.

Time-resolved reflectivity experiments were performed at room temperature by using a passively mode-locked Ti:sapphire laser delivering 50-fs pulses at a repetition rate of 82 MHz. The setup consists of a standard pump probe scheme with orthogonally polarized pump and probe pulses focused on a common spot of 45 μm on the sample. The pump (probe) powers were adjusted to 70 mW (2 mW). The excitation density is estimated to be 7.8×10^{17} electron-hole pairs per cm^3. The coherent phonons were also recorded at different laser energies varied around the first fundamental interband transitions of the Fibonacci superlattices. The interband transition energies of the samples were obtained from photoreflectance experiments.[12] For a high-sensitivity data acquisition we combined a fast-scan detection system,[8] where the time delay was achieved via a speaker scanner operating at 68 Hz, with a stepper delay stage. Several high-resolution data in the 10-ps delay range were combined to long-delay-time scans (>100 ps). The oscillations were numerically ex-

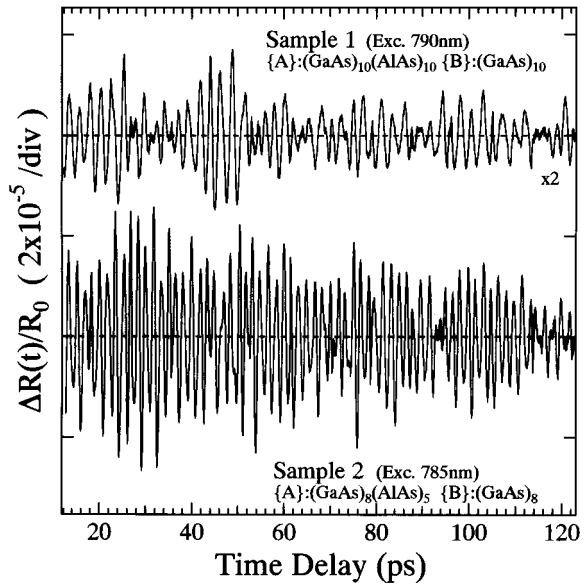

FIG. 1. Oscillatory parts extracted from time-resolved reflectivity changes of Fibonacci superlattices at room temperature.

tracted by subtracting a slowly varying background resulting from bleaching of the optical transitions.

Figure 1 shows the time-resolved oscillatory reflectivity changes of samples 1 and 2 excited at laser wavelengths of 790 nm and 785 nm, respectively. The amplitude of the phonon-induced reflectivity change is of the order of $10^{-5}R_0$, where R_0 is the unperturbed reflectivity. The signal-to-noise ratio of the detection system is below $10^{-6}R_0$. It is obvious from Fig. 1 that many beat features and phase jumps are observed in the oscillatory reflectivity changes. The signals are much more complicated than single-frequency-mode oscillations obtained from periodic superlattices.[9] The main characteristic frequencies are 0.42 THz (sample 1) and 0.60 THz (sample 2). The decay time of the oscillations exceeds 70 ps, which is much longer than the decay times of LO phonons (a few picoseconds) in GaAs-based heterostructures.[10] This indicates the difference in the decay channels of optical and acoustic modes. A comparison of the decay times of acoustic modes in the Fibonacci superlattice and in periodic superlattices[9] shows that the decay times are very similar in both systems.

For a detailed frequency analysis of the time-domain data we perform a numerical Fourier transform[13] depicted in Figs. 2(a) and 3(a). Many sharp peaks appear at nonequal intervals in the Fourier spectra in contrast with the single-mode feature in periodic superlattices.[9] Although we obtained the Fourier spectra in the frequency range up to 2 THz, no significant amplitude of coherent phonons with frequency above 1.2 THz is observed. A long delay-time scan of more than 100 ps in time-resolved reflectivity measurements enables us to obtain the fine structures in the phonon spectra. It is expected that these Fourier spectra reflect the quasiperiodicity of the Fibonacci sequence. The quasiperiodicity is given by

$$d_{m,n}=\frac{d_0}{m+n\tau}, \tag{1}$$

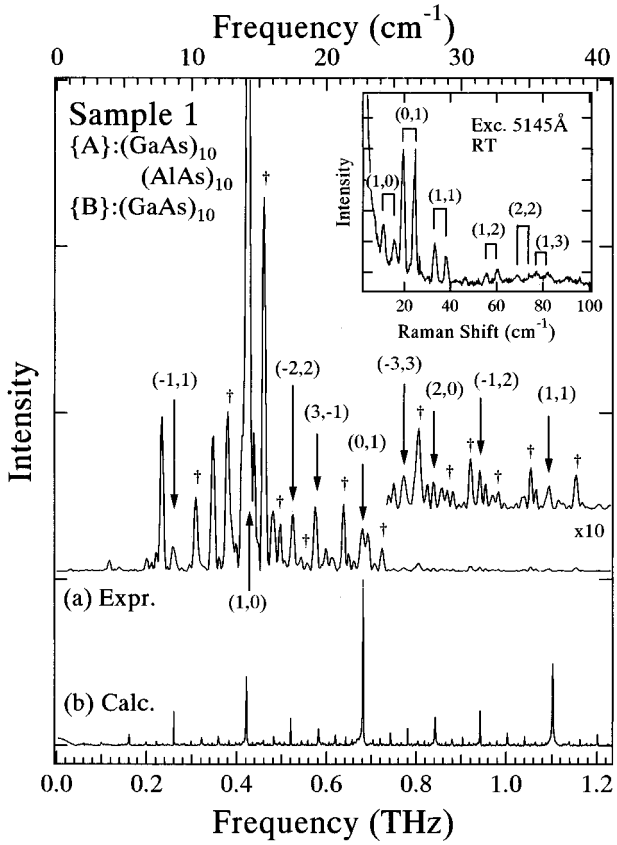

FIG. 2. (a) Fourier spectra of temporal traces in sample 1 shown in Fig. 1. The bottom curve (b) shows calculated spectra $|P_{m,n}|$. The inset shows the Raman spectrum at room temperature with excitation at 514.5 nm.

where m and n are integer indices of the quasiperiodicity, $d_0=\tau d_A+d_B$ is the average lattice parameter, d_A and d_B are the thicknesses of the layer A and B, and $\tau=(1+\sqrt{5})/2$ is the golden mean. On the basis of an elastic continuum model, the characteristic frequency $\omega_{m,n}$ of the folded LA phonon mode labeled by the quasiperiodicity index (m,n) is given by

$$\omega_{m,n}=|k\pm q_{m,n}|v_{SL}, \tag{2}$$

where $q_{m,n}=2\pi/d_{m,n}$, k is the phonon wave vector, and v_{SL} is the effective sound velocity along [001] obtained by simply adding the transit time in each layer (4.726×10^5 and 5.71×10^5 cm/s for GaAs and AlAs, respectively).[7]

The arrows in Figs. 2 and 3 indicate the frequencies of the folded LA phonon modes with $k=0$ calculated from Eqs. (1) and (2) and (m,n) indicates the index of the quasiperiodicity. The most intense band at 0.42 THz (0.60 THz) in the spectrum of sample 1 (sample 2) is attributed to the (1,0) mode. In a comparison of the Fourier spectra [Figs. 2(a) and 3(a)] with the Raman spectra shown in the insets of Figs. 2 and 3,[4] it is obvious that fine structures appear in the Fourier spectra. The most intense bands in the Raman spectra taken with a backscattering geometry are the (0,1) mode, while those bands in the Fourier spectra are the (1,0) mode. In the lower-frequency region of the Fourier spectra, in addition, the tail of the Rayleigh line is not observed as in Raman spectra.

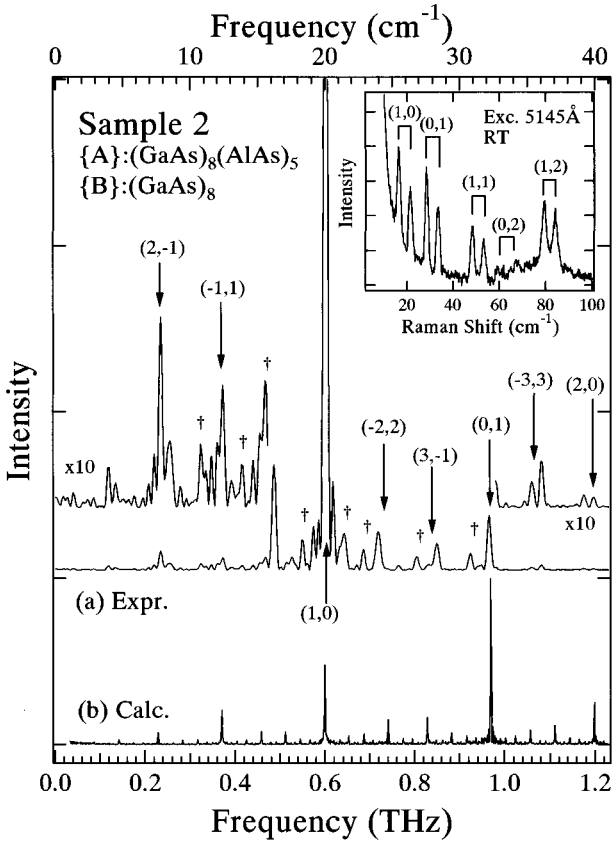

FIG. 3. (a) Fourier spectra of temporal traces in sample 2 shown in Fig. 1. The bottom curve (b) shows calculated spectra $|P_{m,n}|$. The inset shows the Raman spectrum at room temperature with excitation at 514.5 nm.

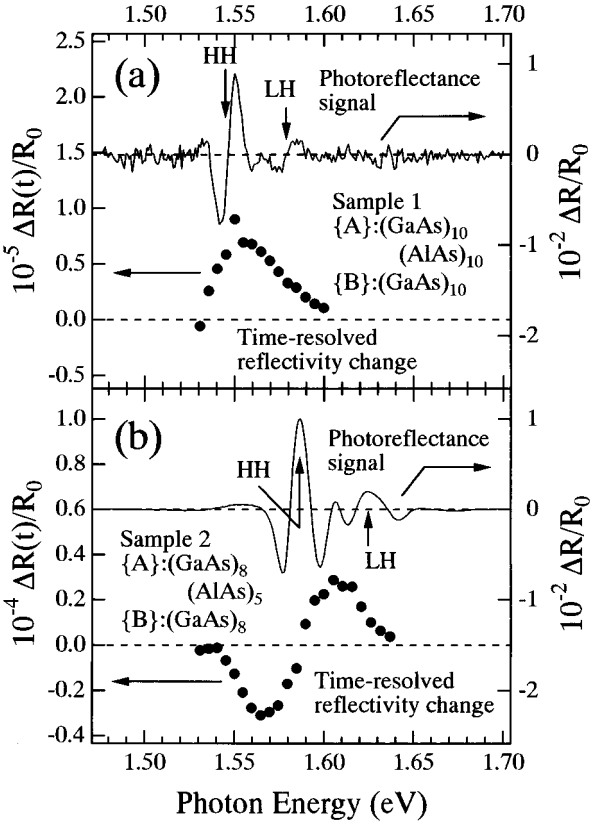

FIG. 4. Comparison of the photoreflectance spectra of (a) sample 1 and (b) sample 2 at room temperature with the energy dependence of the coherent phonon amplitude of the (1,0) mode.

The experimental data are compared to the spectra calculated by using the photoelastic model.[11] This model describes the superlattice as a stack of continuum media with known photoelastic coefficients. The modulation of photoelastic coefficients along the growth direction $z\|[001]$ reflects the Fibonacci sequence in this case. The reflectivity changes obtained from pump-probe measurements are related to the polarizability changes. Based on the photoelastic model, the Fourier component $|P_{m,n}|$ of the spatial modulation of photoelastic coefficients corresponds to the polarizability change due to the (m,n) mode. The curves in Figs. 2(b) and 3(b) show the Fourier spectra $|P_{m,n}|$, where the values of the photoelastic coefficients are 0.48 for GaAs and 0.05 for AlAs.[11] Comparing Figs. 2(a) and 3(a) with Figs. 2(b) and 3(b), we notice that (m,n) modes accompany various peaks. If the Raman process contributes to the generation of the coherent phonons,[14,15] the coherent phonon modes that are shifted to the lower- and higher-frequency sides with kv_{SL} (1.5 cm^{-1}) may appear besides calculated phonon modes with $k=0$. A number of doublet peaks of (m,n) modes as shown by the daggers in Figs. 2(a) and 3(a) can be explained by this assumption.

By comparing the observed spectra with the calculated ones, we see that the intensity profiles disagree with the calculated spectra, while the Raman spectra under a nonresonant condition are explained well by the photoelastic model.[4] It is considered that the coupling between electron and phonon states affects the intensity profile of the coherent phonon. The photoelastic model does not include such a coupling. This is a possible reason for the difference of the intensity profiles. There is another possibility that specific modes are resonantly enhanced in this pump-probe measurement. The Raman spectra were observed under off-resonant conditions at 514.5 nm, while the time-resolved data were obtained using photons resonant with the interband transition. In order to understand the difference between two spectra, it is desirable to compare the coherent phonons in periodic and quasiperiodic superlattices for various excitation photon energies.

For the verification of a resonant process, we investigate the excitation-energy dependence of the amplitude of the dominant (1,0) mode for $k=0$. Figure 4 compares the results obtained from the time-resolved measurements with the photoreflectance spectra of the samples at room temperature. The structures of the photoreflectance spectra originate from the heavy-hole (HH) and light-hole (LH) exciton transitions[12] whose energies are indicated by the arrows in Fig. 4. The splitting of the lowest electron state due to the quasiperiodicity in sample 1 (2) is estimated to be less than 1 meV (about 5 meV) from the calculations based on a transfer-matrix method;[16] therefore, such an energy splitting is hidden by thermal broadening at room temperature. Around the HH-transition energy the coherent acoustic phonon amplitude is strongly modified. The profiles of the excitation-energy dependence of the (1,0)-mode amplitude are broader than the line shapes of the photoreflectance spec-

tra because of the spectral width of the laser pulses of 40 meV and because of carrier-carrier scattering at excitation densities above 10^{17} cm^{-3}. Moreover, in sample 1 only positive amplitudes are observed with a maximum at the HH transition, while the amplitudes in sample 2 exhibit a π-phase shift at the excitation close to the resonance. The difference of the profiles of the excitation-energy dependence between two samples may be caused by the difference of a Fabry-Pérot-type interference effect on the reflectivity peculiar to the layered structures.[9,17] This remarkably modifies the reflectivity change due to exciton transitions as shown by the difference of the photoreflectance line shapes of the two samples in Fig. 4.

In conclusion, we have observed the time-domain spectra of the acoustic phonons in Fibonacci superlattices by pump-probe measurements. The Fourier spectra of the temporal traces of the coherent phonon oscillations show many folded acoustic modes caused by the quasiperiodicity of the Fibonacci sequence. The decay time of the coherent phonons is more than 70 ps, which is comparable to that in periodic superlattices. We demonstrate that a long-delay scan in the time-resolved reflectivity measurements enables us to obtain the fine structures in the Fourier spectra as compared with Raman spectra. The excitation-energy dependence of the temporal traces indicates that the resonance of the HH-exciton-transition remarkably modifies the oscillation amplitude.

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