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Acoustic Modes of GaAs Nanopillars Studied with Ultrashort Optical Pulses

超短光パルスを用いた GaAs ナノピラーの振動モード観測

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1. Introduction

Vibrations of nanostructures can be conveniently probed in a noncontact way in the time domain using ultrashort optical pulses for excitation and detection with typical resonant frequencies in the gigahertz range. Past studies have concerned the observation of individual modes of various nanostructures: nanocubes [1], nanorods [2], and collective modes of two-dimensional periodic structures, for example [1]. Understanding vibrations in nanostructures are important in applications in mass sensing, [3,4] for example.

We have been investigating vibrations of GaAs nanopillars of hexagonal cross section on a GaAs(111)B substrate using an pump-probe technique with ultrashort optical pulses. This work was presented in this symposium last year. Acoustic oscillations are excited by the pump pulses through picosecond stress generation by the thermoelastic and deformation potential mechanisms. For probing, the reflectivity change induced by acoustic oscillation and propagation in the samples is monitored as a function of time [5]. We can measure the frequencies, the attenuations and the phases of the oscillation modes. Moreover, we observed Brillouin oscillations originating in the substrate [6,7]. However, we did not yet determine the shapes of the vibrational modes of the nanopillars.

In this paper, we examine the vibrational properties of GaAs hexagonal nanopillar arrays on a GaAs (111) substrate using a combination of femtosecond pump-probe spectroscopy and theoretical calculations based on the finite element method (FEM).

2. Samples

GaAs nanopillars are prepared on a GaAs (111)B substrate by SA-MOVPE (selective-area metal-organic vapor phase epitaxy) [8], as shown in **Fig. 1**.

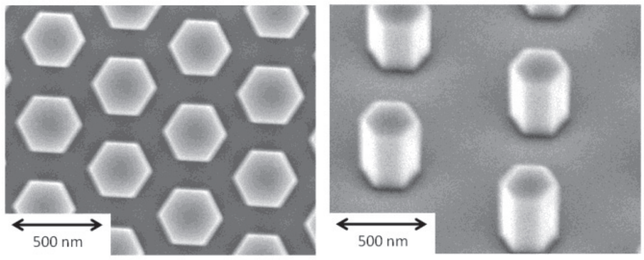

Fig. 1 Typical scanning electron microscope images of GaAs nanopillars. (a) is an image from the top. (b) is an image from an oblique direction.

They have diameter 100-400 nm, height 160-380 nm, period 400-1000 nm, and triangular lattice periodicity. The sizes are estimated from scanning electron microscope images such as **Fig. 1**. These nanopillars are single-crystal, anisotropic and free-standing. We prepared 14 different nanopillar samples in all.

3. Experimental setup

We use the optical pump-probe technique with a mode-locked Ti:Sapphire laser of central wavelength 830 nm, pulse duration ~200 fs, and repetition rate 80 MHz. The laser beam is divided into two, one for the pump light and another for the probe light. The pump beam (wavelength 415 nm) is focused to a ~20 μm diameter spot on the sample using a single lens. The pump pulses generate vibrations of the nanopillars at frequencies up to ~30 GHz through the thermoelastic and deformation potential mechanisms. The probe beam (wavelength 830 nm) passes through a variable delay line, and is focused on the sample also to a ~20 μm diameter spot using a single lens. Probe pulses detect the excited vibrations as reflectivity changes by modulation based on photoelasticity and geometry changes from the nanopillar motion. Both pump and probe beams are normally incident on the samples to simplify the mode excitation and the symmetry of the detection. Typically 1000-5000 pillars are simultaneously probed. Lock-in detection allows us to monitor typical relative reflectivity changes of 10^{-5} order. All the experiments are

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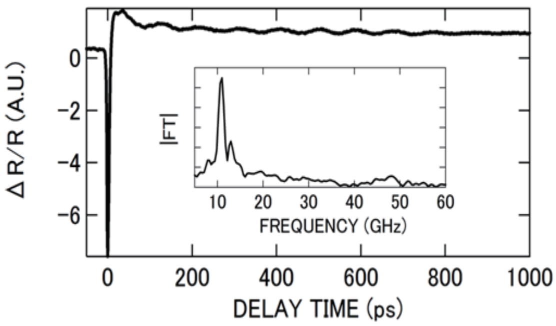

Fig. 2 Relative reflectivity change versus the time delay between the pump and the probe pulses. The nanopillar has a height of 315 nm, a width of 315 nm, and a distance between nanopillars of 1000 nm. The inset shows the vibrational spectrum obtained from the modulus of the temporal Fourier transform of the reflectivity changes.

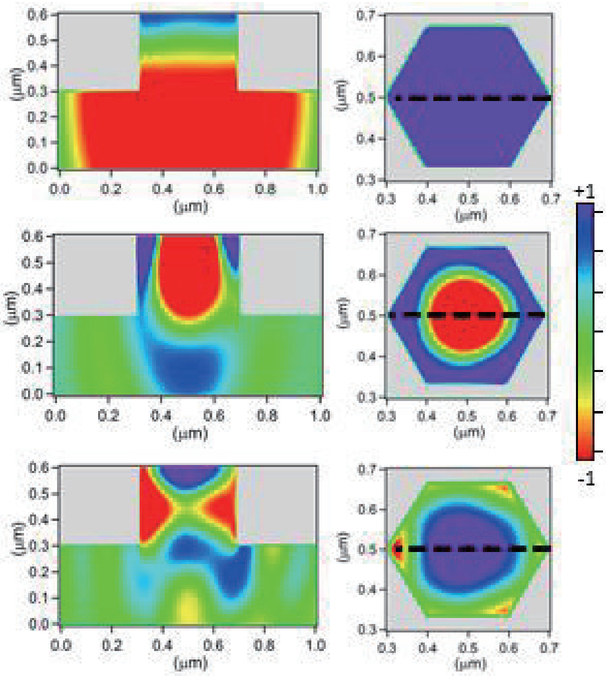

Fig. 3 Images of the *z*-displacement for modes at 2.0, 7.5, 11.0 GHz for nanopillars with height 300 nm, width 330 nm and distance between nanopillars 1000 nm. Left-hand figures are for the cross section, and right-hand figures are for the top surface. Dashed lines indicate the respective cross sections on the left.

performed at room temperature.

4. Results and discussion

We obtained data from 14 kinds of nanopillar sample. A typical experimental result for the relative reflectance change is shown in **Fig. 2**. The fourier spectrum of the reflectivity is shown in the **Fig. 2** inset, showing peaks near 11 GHz and 48 GHz. The peak at 11 GHz is the main vibrational mode of the nanopillars. The peak at 48 GHz corresponds to Brillouin oscillations [6,7].

It has been shown that the main vibrational mode of the pillars is “breathing-like mode” by comparing the experiment with the fundamental infinite cylinder breathing mode [2,6,7]. The frequencies of this dominant mode are inversely proportional to diameter.

More detailed understanding can be achieved using time-domain finite-element simulations. The simulations of 2 sizes of nanopillars done in the experiment are calculated. The modelled periodic array samples are hexagonal, freestanding on the substrate and anisotropic. We use an impulsive vertical external force applied to the top surface of the pillars. We calculate the transient displacement fields of the nanopillars. As the result we obtain by temporal Fourier transforms the shapes of vibrational modes, as shown in the example of **Fig. 3**. In **Fig. 3** we show the vertical displacement (in the *z* direction) of the top surface and for vertical cross sections. There exists three-fold symmetry from the crystal structure in the top surfaces and a left-right asymmetry in the cross section. Collective modes of the two-dimensional periodic structures were not accessible because of their lower frequencies and because of significant vibrational energy losses to the substrate.

5. Conclusions

We have investigated the vibrational modes of GaAs nanopillars of hexagonal cross section on a GaAs substrate using an ultrafast optical pump-probe technique. Breathing modes of the pillars are detected in the 10-30 GHz range. Furthermore, the vibrational modes of the nanopillars have been successfully simulated by finite-element modelling.

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