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Sub-terahertz-frequency attenuation and sound velocity of GaN at Cryogenic Temperatures Studied by Picosecond Ultrasound Spectroscopy

サブ THz 超音波を用いた GaN の音速・減衰係数の極低温計測

Akira Nagakubo[1, 2†], Yuki Tsutsui[1], Hirotsugu Ogi [1], Nobutomo Nakamura[1], and Masahiko Hirao[1] ([1]Grad. School of Engineering Science, Osaka Univ. ; [2] JSPS Research Fellow)

長久保 白 [1, 2†], 筒井 裕貴 [1], 荻 博次 [1], 中村 暢伴 [1], 平尾 雅彦 [1] ([1]阪大大学院 基礎工, [2]学振特別研究員)

1. Introduction

Wurtzite GaN (W-GaN) has a wide band gap of 3.4 eV, corresponding to 365-nm ultraviolet light. It is very useful for many applications such as laser diodes and light emitting diodes (LED). Moreover, W-GaN and its alloys are piezoelectric materials with large piezoelectric constants.

The Q value of a piezoelectric material is very important because it determines the energy loss of the material. It reflects principally the phonon-phonon interaction and also the interactions between point defects and dislocations. Therefore, internal-friction measurements can reveal such internal phonon behaviors. Especially, a high-frequency and short-wavelength ultrasound is expected to be more sensitive for such behaviors. Although W-GaN is a candidate of acoustic resonator, the difficulties of synthesis have prevented accurate measurements of acoustic characteristics. Recently, we measured the all independent elastic constants and piezoelectric coefficients of W-GaN at room temperature by resonance ultrasound spectroscopy with laser-Doppler interferometory[1)]. However, intrinsic elastic properties are still unclear. For example, the temperature dependences of elastic moduli reflect anharmonicity of the inter-atomic potential. High-temperature behaviors can contribute to designing applications and cryogenic-temperature behaviors reveal important physical properties relating to phonon vibrations. For designing oscillators, it is also important to measure attenuation behaviors at high frequency.

Previous works applied picosecond ultrasound method to W-GaN[2,3)]. However, they failed to measure attenuation and low temperature elasticity. Then, we measure the attenuation and sound velocity of GaN single crystals by picosecond ultrasound spectroscopy[4,5)] between room temperature and cryogenic temperature. We adopt a long-delay line (~2.6 ns) and use a short-wavelength probe light to observe higher-frequency phonons excited by a thin film's thermal expansion deposited on a specimen. Low-temperature measurements contribute to comparison of calculation results and experimental results because most first-principle calculations neglect phonon vibration and their results correspond to the physical properties at cryogenic temperature. The synthesis of GaN has difficulties and calculational prediction will have some role.

2. Experiments

Picosecond ultrasound spectroscopy uses an ultrafast pump light pulse to excite an ultrasound and a probe light pulse to detect it. We developed an optics for a long-time-delay and cryogenic temperature measurements, as shown in **Fig. 1**. This optics has a 200-mm delay line equipped two corner reflectors. We use a titanium-sapphire pulse laser and divided the source light pulse into pump and probe light pulses by a polarization beam splitter (PBS). Their power ratio is adjusted by a $\lambda/2$ wave plate.

The pump light is modulated at 100 kHz by an acousto-optical (AO) crystal, and the probe light's frequency is doubled by a second harmonic generator (SHG) crystal. Both of them are perpendicularly incident on a specimen and we distinguish them by a sharp color-cut dichroic mirror (DM), which reflects 800-nm-wavelength light and transmits 400-nm-wavelength light.

We deposited a 10-nm Al thin film on the specimen as a sound generator through the ultrafast thermal expansion and shrinking caused by the pump light. The ultrasound works as a diffraction grating for the probe light. Backward diffracted probe light and reflected probe light at the surface interfere with each other. We can observe an oscillating signal in the reflectivity as the ultrasound propagates. This is Brillouin oscillation, and its frequency f is well approximated by the Bragg's condition of backward diffraction[5)], that is

$$f = \frac{2nv}{\lambda} \qquad (1)$$

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ogi@me.es.osaka-u.ac.jp

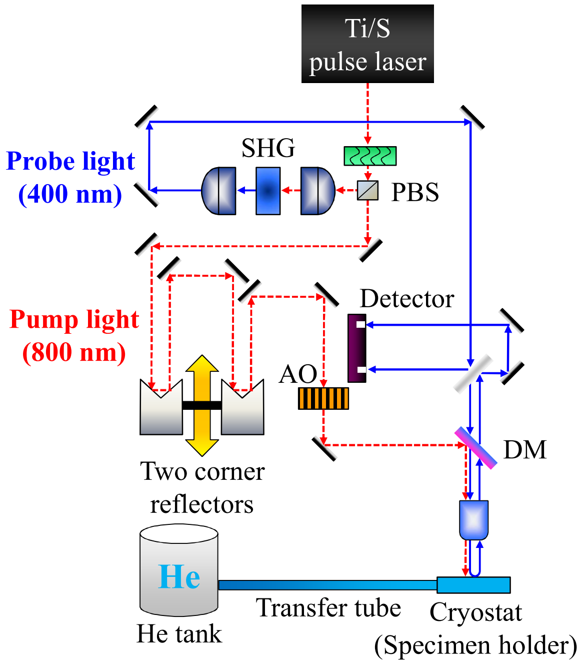

Fig. 1 Schematic of the optics. Solid and dashed lines show 400-nm and 800-nm wavelength light, respectively.

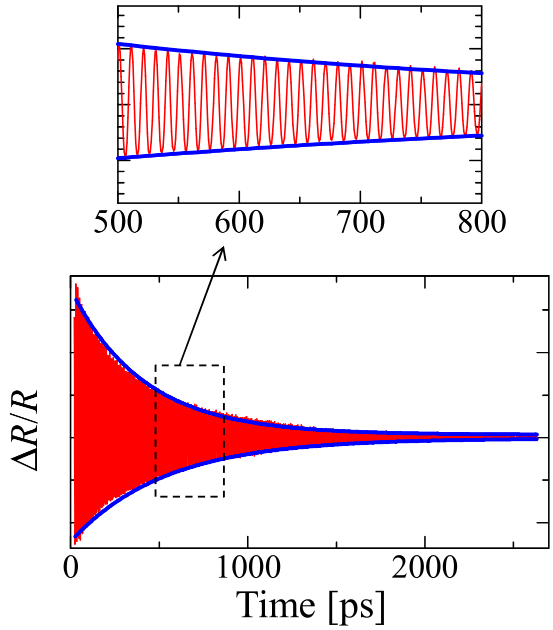

Fig. 2 Brillouin oscillation in GaN. The enveloped curves are fitted by exponential.

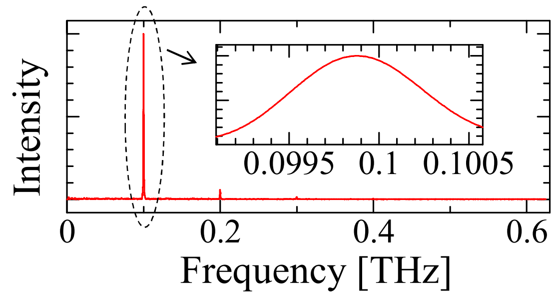

Fig. 3 The FFT spectrum of the Brillouin oscillation at room temperature. A small-second harmonic component is observed.

where n is the refractive index, v is the longitudinal sound velocity, and λ is the wavelength of the probe light. We determine attenuation coefficients by fitting exponential function to the damping oscillation envelope.

3. Results and Discussions

At room temperature, we succeeded in observing high amplitude Brillouin oscillations as shown in **Fig. 2**. A clearly damping oscillation is observed, and we determined attenuation coefficient α to be 2.15 ns^{-1} at room temperature. **Figure 3** shows the fast Fourier transform. Measured sound velocity and elastic constant of C_{11} are slightly higher than the reported value measured resonance ultrasound spectroscopy method[1], indicating the piezoelectric stiffening effect.

Attenuation naturaly decreases at low temperature. On the other hand, the temperature dependence of the Brillouin oscillation frequency has a positive sign. This is very interesting because most material's elastic constants and Brillouin oscillation frequency show negative temperature dependences. It would reflect the temperature dependence of the refractive index[6], and we are now carefully investigating this reason.

Acknowledgement

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