Accurate Measurement of Nonlinear Optical Coefficients of Gallium Nitride

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1. Introduction

III-nitride semiconductors, especially gallium nitride (GaN), aluminum nitride (AlN), and their compounds (AlGaN), are promising materials for semiconductor photonic devices operating in the ultraviolet wavelength region because of their wide band gaps (3.4 eV for GaN and 6.2 eV for AlN). Although the laser diodes operating at 342 nm are now realized using AlGaN, those at shorter wavelength have not been reported. Wavelength conversion devices using AlGaN as a nonlinear optical material monolithically integrated with nitride laser diodes can be a coherent ultraviolet light source. Nevertheless the accuracies of the reported values of quadratic nonlinear optical coefficients of the incident fundamental and the output second-harmonic (SH) waves.

2. Experiment

We first performed rotational Maker-fringe measurement on HVPE-grown GaN. We prepared a 275-µm-thick (0001) plane-parallel plate of GaN by polishing the both surfaces of a HVPE-grown bulk GaN sample. Experimental setup was a standard one similar to that described in Refs. [4, 5]. We used a cw-pumped Q-switched Nd:YAG laser (Spectron, SL902) oscillating at 1.064 µm as a fundamental light source. The pulse width was 100 ns, the peak power was 5 kW, and the repetition rate was 5 kHz. The fundamental beam was incident upon the sample as a linearly polarized collimated beam with a radius of 655 µm. We measured the three independent nonlinear-optical coefficients (d₁₁ = d₂₂ = d₁₅ = d₂₅, d₃₃), which were all the non-zero components deduced from the GaN’s hexagonal crystal structure, by changing the polarization of the incident fundamental and the output second-

Figure 1: Maker fringes of an HVPE-grown GaN sample obtained with the (a) s-p (b) p-p (c) 45°-s configurations.
least-squares fitting routine. All the experimental data can be closely fitted with the following set of fitting parameters: $L = 274.8 \mu m$, $n_{e1} = 2.2671$, $n_{e2} = 2.3008$, $n_{o1} = 2.3612$, $n_{o2} = 2.4003$, and the misorientation angle $\Delta \theta = 0.537^\circ$. Although these values of refractive indices are smaller than the values reported by Sanford et al. $[7]$ ($n_{e1} = 2.304$, $n_{e2} = 2.342$, $n_{o1} = 2.397$, and $n_{o2} = 2.435$), we believe these values are highly reliable as accurate refractive indices of the present high-purity GaN sample. Based on the fitting combined with relative measurements using quartz ($d_{11} = 0.30 \text{pm/V} [6, 8]$) as a reference material, we determined the magnitudes of the nonlinear optical coefficients of HVPE-grown GaN as follows: $d_{31} = 2.5 \pm 0.1 \text{pm/V}$, $d_{15} = 2.4 \pm 0.1 \text{pm/V}$, $d_{33} = -7.0 \pm 3.5 \text{pm/V}$. Relatively large error of $d_{33}$ coefficient is due to the small contribution of this coefficient expressed approximately with a projection factor of $\sin^2 \theta$, where $\theta$ is the internal refraction angle, becomes small owing to high refractive indices.

**LPE-grown GaN**

We then performed the wedge measurement on LPE-grown GaN. We obtained a (1120)-oriented plate by cutting a 0.3-mm-thick (0001) plane parallel plate of bulk single-crystalline GaN grown by LPE using the Na flux method $[9]$ perpendicularly to the [1120] direction. Both the input and output facets of the plate, the sizes of which were 0.3\times5 \text{mm}^2, were then polished to the center thickness of $\sim300 \mu m$ and tapered with the apex angles of 0.10 degree. The experimental setup for the wedge measurements was similar to that used in the rotational Maker-fringe measurements except that the fundamental beam was focused onto the sample with a radius of 37.0 \mu m in order to ensure the resolution of Fabry-Perot patterns.

The measured SH power as a function of the sample thickness obtained with a fundamental input polarized perpendicular to the [0001] axis and SH output polarized parallel to the [0001] axis ($d_{33}$ configuration), a fundamental input polarized parallel to the [0001] axis and SH output polarized parallel to the [0001] axis ($d_{13}$ configuration), and a fundamental input polarized at 45° from the [0001] axis and SH output polarized perpendicular to the [0001] axis ($d_{15}$ configuration) are shown in Fig. 2. The short-period oscillations of the SH power superimposed upon the sine-squared Maker fringes were caused by the multiple-reflection effects.

We analyzed the experimental data fully taking account of the multiple-reflection effects $[4, 6]$. The refractive indices of the samples used in the analyses were those determined for HVPE-grown GaN previously. The open circles are the experimental data, while the solid curves show the theoretical curves least-squares-fitted to the experimental data. The agreement between the experiment and theory is quite satisfactory, demonstrating the reliability of our measurement and analysis and excellent quality of our sample. The obtained values of nonlinear optical coefficients are as follows: $d_{31} = 2.3 \pm 0.1 \text{pm/V}$, $d_{15} = 2.6 \pm 0.1 \text{pm/V}$, $d_{33} = -3.8 \pm 0.2 \text{pm/V}$. By virtue of the sample orientation, experimental accuracy of $d_{33}$ measured on the (1120) sample is much higher than that for the (0001) sample.

Since the obtained values of $d_{31}$ and $d_{15}$ coefficients for HVPE- and LPE-grown GaN, which are determined with sufficient accuracies for both samples, agree well within the experimental accuracy, we are convinced that highly accurate values of quadratic nonlinear optical coefficients of GaN are determined by precise measurements on state-of-the-art high-quality samples.

**3. Conclusion**

We have determined all the three independent components of quadratic nonlinear optical coefficients of GaN using high quality samples as follows: $d_{31} = 2.4 \pm 0.1 \text{pm/V}$, $d_{15} = 2.5 \pm 0.1 \text{pm/V}$, $d_{33} = -3.8 \pm 0.2 \text{pm/V}$, where the $d_{31}$ and $d_{15}$ values are averaged ones of those obtained for two samples, and we adopt $d_{33}$ value determined using the (1120) sample that offers much higher accuracies then the (0001) sample. These nonlinear optical coefficients are larger than those of conventional ultraviolet wavelength conversion crystals.

**References**