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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, which determine the efficiency of wavelength conversion devices, of GaN [1, 2, 3] seem to be quite low mainly because of low crystal qualities of samples used in the measurements. In this paper, we report Maker-fringe measurements on high-quality bulk single crystalline samples of GaN and accurate determination of nonlinear optical coefficients taking account of multiple reflection effects.

2. Experiment

We performed Maker-fringe measurement on HVPE- and LPE-grown bulk GaN samples.

HVPE-grown GaN

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. 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 ($d_{31} = d_{32}, d_{15} = d_{24}, d_{33}$), which were all the non-zero components deduced from the GaN's hexagonal crystal structure, by changing the configuration of the polarizations of the incident fundamental and the output second-harmonic (SH) waves.

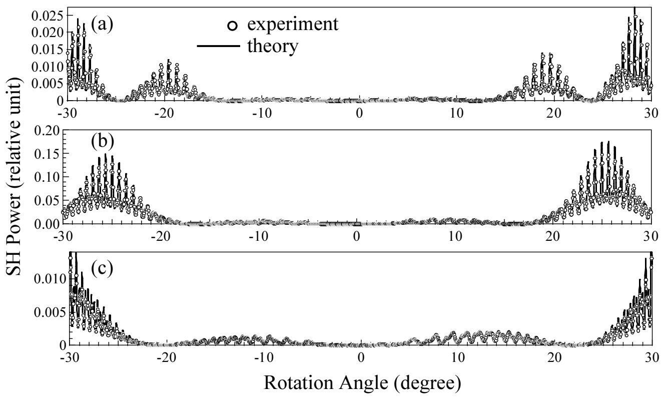

Figure 1: Maker fringes of an HVPE-grown GaN sample obtained with the (a) s-p (b) p-p (c) 45°-s configurations.

Maker fringes of HVPE-grown GaN obtained with an s-polarized fundamental input and a p-polarized SH output (s-p configuration), a p-polarized fundamental input and a p-polarized SH output (p-p configuration), and 45°-polarized (1:1 in-phase combination of s and p polarizations) fundamental input and an s-polarized SH output (45°-s configuration) are shown in Fig. 1. The SH powers are normalized with respect to the envelope of the Maker fringes obtained from a standard sample (an anti-reflection coated *b*-cut quartz plate). As is evident in the data shown in the figures, the Maker fringes are modulated with large amplitude double-period oscillations which are caused by the multiple reflections and interferences of the fundamental and SH waves. Clear Fabry-Perot patterns of the obtained Maker-fringe data indicates an extremely high optical quality of the sample and high accuracies of the experiments. The slight asymmetries of the Maker fringes are caused by an accidental tilt of the plane direction of the sample.

We fitted the experimental data with the theoretical calculations taking account of the multiple-reflection effects [5] and misorientation of the plane direction using a standard

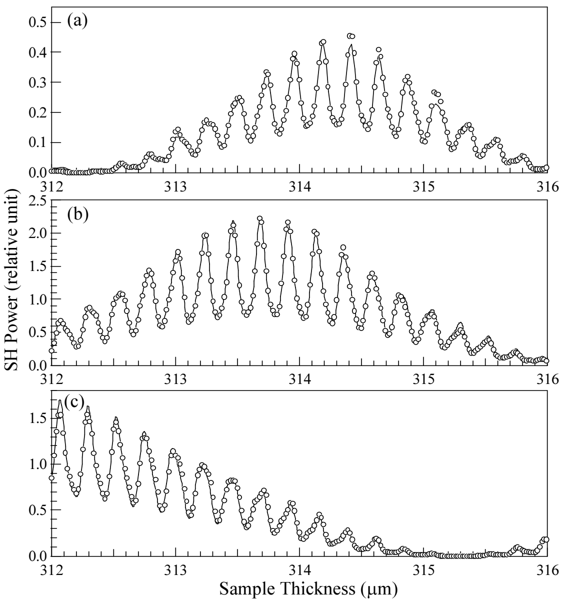

Figure 2: Maker fringes of LPE-grown GaN obtained with the (a) d_{31} (b) d_{33} (c) d_{15} configurations.

least-squares fitting routine. All the experimental data can be closely fitted with the following set of fitting parameters: $L = 274.8$ μm, $n_o^{\omega} = 2.2671$, $n_e^{\omega} = 2.3008$, $n_o^{2\omega} = 2.3612$, $n_e^{2\omega} = 2.4003$, and the misorientation angle $\Delta\theta = 0.537°$. Although these values of refractive indices are smaller than the values reported by Sanford et al. [7] ($n_o^{\omega} = 2.304$, $n_e^{\omega} = 2.342$, $n_o^{2\omega} = 2.397$, and $n_e^{2\omega} = 2.435$), 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 ($d_{11} = 0.30$ pm/V [6, 8]) as a reference material, we determined the magnitudes of the nonlinear optical coefficients of HVPE-grown GaN as follows: $d_{31} = 2.5 \pm 0.1$ pm/V, $d_{15} = 2.4 \pm 0.1$ pm/V, $d_{33} = -7.0 \pm 3.5$ 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 $\sin^3\theta'$, where θ' 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 $(11\bar{2}0)$-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 $[11\bar{2}0]$ direction. Both the input and output facets of the plate, the sizes of which were 0.3×5 mm^2, were then polished to the center thickness of ∼300 μm 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 μm 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_{31} configuration), a fundamental input polarized parallel to the [0001] axis and SH output polarized parallel to the [0001] axis (d_{33} configuration), and a fundamental input polarized 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: $d_{31} = 2.3 \pm 0.1$ pm/V, $d_{15} = 2.6 \pm 0.1$ pm/V, $d_{33} = -3.8 \pm 0.2$ pm/V. By virtue of the sample orientation, experimental accuracy of d_{33} measured on the $(11\bar{2}0)$ 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 the 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: $d_{31} = 2.4 \pm 0.1$ pm/V, $d_{15} = 2.5 \pm 0.1$ pm/V, $d_{33} = -3.8 \pm 0.2$ 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 $(11\bar{2}0)$ sample that offers much higher accuracies then the (0001) sample. These nonlinear optical coefficients are larger than those of conventional ultraviolet wavelength conversion crystals.

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