Second harmonic generation from photonic structured GaN nanowalls

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Abstract. We observed large enhancement of reflected second harmonic generation (SHG) using the one-dimensional photonic effect in regularly arranged InGaN/GaN single-quantum-well nanowalls. Using the effect when both fundamental and SH resonate with the photonic mode, we obtained enhancement of about 40 times compared with conditions far from resonance.

1. Introduction
Second-order susceptibilities $\chi^{(2)}$ of GaN and InGaN are higher than those of conventional second harmonic (SH) crystals such as KDP and LiNbO$_3$ [1]. Nevertheless, the efficiency of SH generation (SHG) in a bulk GaN or InGaN/GaN quantum-well (QW) sample is very small because these are highly dispersive materials, preventing fulfillment of the phase-matching condition.

To increase the SH intensity, we use the photonic effect in an InGaN/GaN single-quantum-well (SQW) nanowall. A nanowall is a plate-like crystal with a thickness of several hundred nanometers and height of a few micrometers [2]. Regularly arrayed nanowalls are considered to be a one-dimensional photonic crystal (1-D PC). We have observed significantly enhanced SHG when both the fundamental and generated SH field are resonant with photonic eigenstates.

2. Principle of photonic enhancement in SHG
A schematic of the experimental configuration is shown in figure 1. A femtosecond pulse (fundamental frequency: $\omega$) was irradiated on the upper surface of the sample along the x axis, and an SH beam (2$\omega$) contained in the reflected fundamental was detected. The direction of the fundamental is specified by the incident angle $\theta$. The principle of SH enhancement is as follows. There are three situations. First, if the in-plane wave vector of fundamental light [$\mathbf{k}_f(\omega) = \mathbf{k}(\omega)\sin\theta$] matches the wave vector of the 1-D PC mode in the nanowall, the fundamental EM field is strongly confined within the PC. Second, if the in-plane wave vector of the generated SH light [$\mathbf{k}_g(2\omega) = \mathbf{k}(2\omega)\sin\theta$] matches that of the 1-D PC mode, the SH light can be efficiently coupled back in the surrounding vacuum. Third, the previous two resonance conditions synchronize,
which causes significant enhancement. These three situations are called the resonant-nonresonant (R-NR), nonresonant-resonant (NR-R), and resonant-resonant (R-R) conditions, respectively [3,4].

Cowan et al. have theoretically shown about $10^6$-fold enhancement in a 2-D GaAs PC slab compared to an untextured sample [3]. Moreover, Torres et al. have experimentally obtained about $10^5$-fold enhancement using a 1-D GaN PC slab [5].

When the crystal c axis is the z direction, considering the crystallite symmetry of GaN and InGaN (class of crystal symmetry 6mm), the induced polarization inside the nanowall is taken as

\[
P_x(2\omega) = 2\varepsilon_0\chi^{(2)}_{xxz}(\omega)E_x(\omega)E_z(\omega)
\]

\[
P_y(2\omega) = 2\varepsilon_0\chi^{(2)}_{yyz}(\omega)E_y(\omega)E_z(\omega)
\]

\[
P_z(2\omega) = \varepsilon_0\chi^{(2)}_{zzz}(\omega)^2 + \varepsilon_0\chi^{(2)}_{zzy}(\omega)^2 + \varepsilon_0\chi^{(2)}_{zxy}(\omega)^2
\]

where $E_i(\omega)$ is the i-component of the fundamental electric field. Equation (1) shows that SH is generated with p polarization when the fundamental was irradiated on the sample with either p or s polarization.

3. Experimental set-up

Figure 2 shows top view and bird’s-eye view scanning electron microscopy (SEM) images of our nanowall sample. The nanowall’s periodicity and width are 504 nm and 236 nm, respectively. The patterned area is 150 x 150 \( \mu \)m\(^2\). Pulses (pulse width \( \approx 100 \) fs) from a mode-locked Ti:sapphire laser were used for fundamental light and focused onto the sample with a spot size of \( \sim 180 \) \( \mu \)m. The polarization of the fundamental and SH was determined using a half-wavelength plate and a polarizer. The angle of incidence was fixed, and the fundamental wavelength dependency of the SH intensity was measured in the range from about 770 nm to 915 nm.

4. Experimental result

4.1. R-NR condition

Figure 3 shows an example of the photonic band structure (PBS) (\( \theta = 20^\circ \)) calculated by the transfer matrix method. Because the InGaN layer is thin (a few nanometer wide), the PBS was calculated assuming that the nanowall was composed only of GaN. The refractive index dispersion of GaN was used in the calculation. The vertical axis is the normalized frequency, and the horizontal axis is the
in-plane wavenumber $k_y = k \sin \theta$. The squares show the PC mode for the $s$-polarized fundamental. The solid line shows the dispersion of light in vacuum in the case of $\theta = 20^\circ$. The SH intensity is resonantly enhanced when the dispersion of incident light crosses the squares. This corresponds to the R-NR condition. In this case, we can expect an increase in SH intensity at 2.85 eV.

Figure 4 shows the observed SH intensity at $\theta = 20^\circ$. The fundamental was $s$-polarized, and a $p$-polarized SH signal was detected. We observed a clear enhancement of SH intensity at 2.85 eV.

4.2. NR-R and R-R conditions

Figure 5 shows the PBS in the case of $\theta = 23.4^\circ$. The squares show the PC mode for the $s$-polarized fundamental. The triangles show the $p$-polarized mode plotted at half the frequency and in-plane wavenumber; therefore, they show the points where the generated SH beam resonates with the PC mode. The solid line shows the dispersion of light in vacuum in the case of $\theta = 23.4^\circ$. The SH intensity is resonantly enhanced when the dispersion of incident light crosses the squares and/or the triangles. In this case, we can expect an increase at 2.92 eV (NR-R condition) and substantial enhancement at 2.78 eV (R-R condition).
Figure 6 shows the observed SH intensity at $\theta = 23.4^\circ$. We observed a small enhancement of SH intensity at 2.92 eV and a significant enhancement at 2.78 eV. At the R-R point, the degree of SH enhancement was about 40 times that of about 3 eV, far from the resonance.

Figure 7 shows the fundamental polarization dependency of SH intensity in the R-R condition. SH intensity decreased remarkably when the polarization of the fundamental was changed to $p$ polarization, because the R-R condition is satisfied only for the fundamental light with $s$ polarization.

5. Discussion
At first glance, it seems that our enhancement factor of ~40 is much smaller than that observed by Torres et al. [4]. However, our enhancement factor is not a comparison between a film and a nanowall, but between the R-R point and a nonresonant point. There may be an enhancement even at the nonresonant point, because the wavelength region measured is not significantly away from the resonance point. In fact, Torres et al. also measured the change in SH intensity as a function of the fundamental wavelength [6], and their result is almost the same as ours.

It should be mentioned that the upper surface of our sample is not flat (figure 2), and thus, the generated SH is diffused because of scattering. Therefore, we could not detect all of the generated SH. In fact, the reflected fundamental light of the nanowall was more extensive than that of our film sample. Of course, imperfection of the period of the nanowall also influences the enhancement factor.

Several improvements could be made in our sample to obtain a stronger SH signal. If the upper surface of the nanowall is flat, a greater amount of the fundamental will be incident on the PC core, and the influence of scattering will be smaller in the radiation process of the SH beam. In addition, Cowan et al. showed that SH intensity is also influenced by the quality factor $Q$ of the resonant mode [3], which is decided by the sample’s structure in the $z$ direction. Progress in controlling the growth of nanowalls will lead to achieving a perfect period and the design of nanowalls with a high $Q$.

6. Conclusion
We have observed enhancement of reflected SH intensity using the 1-D PC effect in an InGaN/GaN SQW nanowall. By calculating the PBS, we found the R-R point at which both the fundamental and SH were resonant with the PC mode. In the R-R condition, we observed significantly enhanced SH signal. There is still great potential for further increasing the SH intensity by improving the sample structure.

References
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