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An attenuated-total-reflection study on the surface phonon–polariton in GaN

K Torii†, T Koga†, T Sota†¶, T Azuhata‡, S F Chichibu§ and S Nakamura‖+

† Department of Electric, Electronics, and Computer Engineering, Waseda University, Shinjuku, Tokyo 169-8555, Japan
‡ Department of Materials Science and Technology, Hirosaki University, Hirosaki, Aomori 036-8561, Japan
§ Institute of Applied Physics, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8573, Japan
‖ Department of Research and Development, Nichia Chemical Industries Limited, Oka, Kaminaka, Anan, Tokushima 744-8601, Japan

E-mail: kousuke@mn.waseda.ac.jp (K Torii) and tkyksota@mn.waseda.ac.jp (T Sota)

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Abstract. A study on the surface phonon–polariton in wurtzite GaN has been carried out. Polarized attenuated-total-reflection spectra reveal that an absorption peak appears at 699 cm^{-1} only for the incident beam with P polarization. Theoretical calculations demonstrate that the observed absorption is due to the surface phonon–polariton.

It is known that infrared-active (IR-active) transverse mode lattice oscillations in materials couple with electromagnetic waves to produce the elementary excitation called the phonon–polariton in the long-wavelength region. The nature of the phonon–polariton is in part photon and in part phonon. The surface phonon–polariton is defined as a phonon–polariton that travels along a direction perpendicular to the surface normal, and its amplitude attenuates in going from surface to bulk. The surface phonon–polariton is a transverse magnetic (TM) mode vibration [1–3].

To observe the surface phonon–polariton, measurements using Raman scattering and/or attenuated total reflection in the Otto configuration [4, 5] are usually made [1–3]. Using Raman scattering, Davydov *et al* studied surface and interface phonon–polaritons in a cubic GaN epitaxial layer grown on a (001) GaAs substrate [6]. The ATR method uses evanescent waves which can directly excite the surface phonon–polariton [1, 2, 4, 5]. Because of this merit, the ATR method is widely used to study the surface phonon–polariton in materials [1, 2]. However, to the best of our knowledge, there is no publication in which the ATR method has been applied to study the surface phonon–polariton in GaN.

In this work, we report on the observation of the surface phonon–polariton in free-standing bulk GaN with a wurtzite structure using the ATR method in the Otto configuration [4, 5]. The observed signal is compared with theoretical calculations. A good agreement between them is obtained.

¶ Also with: Materials Research Laboratory for Bioscience and Photonics, Graduate School of Science and Engineering, Waseda University, Japan.
+ Present address: Materials Department, University of California, Santa Barbara, CA 93106, USA.

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The sample used here was a free-standing *c*-face GaN bulk sample of high quality. The sample was grown by the two-flow metal–organic chemical vapour deposition method [7] using the lateral epitaxial overgrowth technique [8–10]. The sample size was about 5 mm × 10 mm × 70 μm. The characteristics of the sample have been published elsewhere [11, 12].

ATR spectra were measured using a JASCO FT/IR-430 attached with an optional ATR unit at room temperature. We used a KRS-5 (Kristall aus des Schmeizfluss (TlBr 44% + TlI 56%)) as a prism, which makes it possible to measure ATR signals in the region of frequencies larger than 500 cm^{-1}. The resolution was 1 cm^{-1}. The total-reflection angle of the IR beam in the prism was about 45°. To realize the Otto configuration, gold leaves are inserted into the interface between the sample and the prism avoiding the IR beam path. The width of the air gap was set as about 1 μm. A wire grid polarizer was used to obtain the linear polarization beam.

The IR beam with S polarization, i.e., with the electric field direction associated with the IR beam parallel to the sample surface, may excite transverse electric (TE) mode vibration. The IR beam with P polarization, i.e., with the electric field direction perpendicular to that in the case of S polarization, may excite TM mode vibration. In the right-hand-side panel of figure 1 are shown polarized absorption spectra obtained experimentally. It is found that

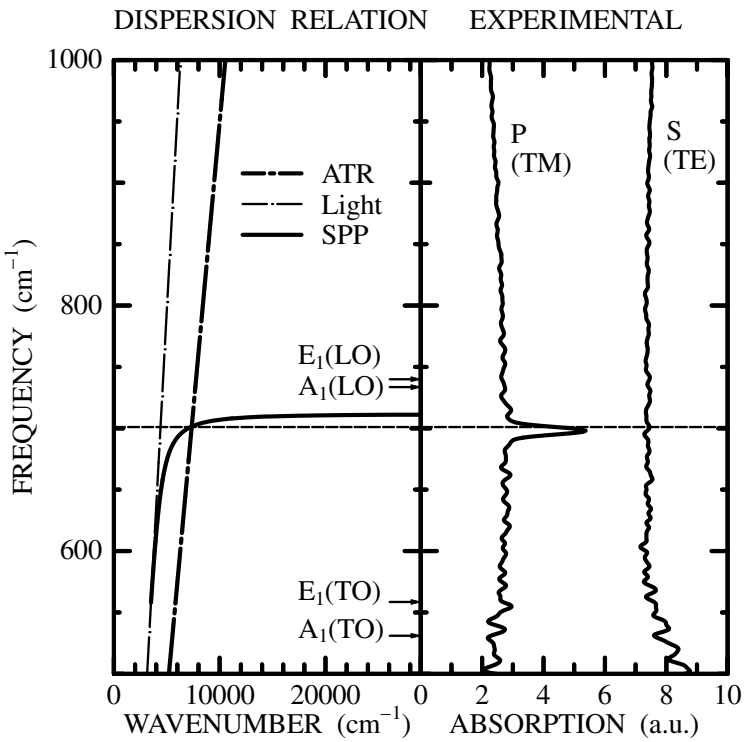

Figure 1. Polarized absorption spectra obtained from the ATR measurements on wurtzite GaN are shown in the right-hand-side panel. The letters S and P indicate the polarization of the incident beam. TE and TM are abbreviations for the transverse electric and transverse magnetic modes. The incident beam with S (P) polarization excites the TE (TM) mode vibration. In the left-hand-side panel, the following theoretically calculated curves and lines are shown. The solid curve labelled SPP is the dispersion relation for the surface phonon–polariton. The light and ATR scan lines are, respectively, shown by the thin and thick chain lines. The frequency positions of $\omega_{\mathrm{TO,A_1}}$, $\omega_{\mathrm{TO,E_1}}$, $\omega_{\mathrm{LO,A_1}}$, and $\omega_{\mathrm{LO,E_1}}$ are also shown.

no absorption is observed for the S-polarized beam, while an absorption peak is observed at 699 cm^{-1} for the P-polarized beam. This selection rule suggests that the observed peak is attributable to the absorption due to the surface phonon–polariton.

To confirm the above assignment, theoretical calculations have been carried out. We compare the experimentally obtained absorption peak position and the intersection between the dispersion relation for the surface phonon–polariton in GaN and the ATR scan line determined by the refractive index and the shape of the prism used. According to Mirlin [1], an implicit dispersion relation for the surface phonon–polariton propagating on the *c*-face of a material with a point group of C_{6v} is given by

$$\frac{q^2c^2}{\omega^2} = \varepsilon_{\parallel}(\omega)\frac{1-\varepsilon_{\perp}(\omega)}{1-\varepsilon_{\perp}(\omega)\varepsilon_{\parallel}(\omega)}. \tag{1}$$

Here ω and c are, respectively, the angular frequency of the surface polariton and the velocity of light, q is the absolute value of the wave vector $\boldsymbol{q}$ of the surface phonon–polariton, and $\varepsilon_{\parallel}(\omega)$ ($\varepsilon_{\perp}(\omega)$) is the dielectric function parallel (perpendicular) to the *c*-axis. Assuming that the phonon lifetime is infinite for simplicity, the concrete expressions for $\varepsilon_{\parallel}(\omega)$ and $\varepsilon_{\perp}(\omega)$ are as follows:

$$\varepsilon_{\parallel}(\omega) = \varepsilon_{\infty,\parallel}\frac{\omega^2_{\mathrm{LO,A_1}} - \omega^2}{\omega^2_{\mathrm{TO,A_1}} - \omega^2} \tag{2}$$

$$\varepsilon_{\perp}(\omega) = \varepsilon_{\infty,\perp}\frac{\omega^2_{\mathrm{LO,E_1}} - \omega^2}{\omega^2_{\mathrm{TO,E_1}} - \omega^2} \tag{3}$$

where $\varepsilon_{\infty,\parallel(\perp)}$ is the high-frequency dielectric constant parallel (perpendicular) to the *c*-axis and $\omega_{\mathrm{TO,X}}$ and $\omega_{\mathrm{LO,X}}$ ($\mathrm{X} = \mathrm{A_1}, \mathrm{E_1}$) are, respectively, the pure TO and LO phonon frequencies of the X mode.

The dispersion curve calculated using equations (1)–(3) is shown by a solid curve labelled SPP in the left-hand-side panel of figure 1, where the light line in the air is also shown by a thin chain line. The physical parameters used herein are as follows. We set $\varepsilon_{\infty,\perp} = 5.76$ and $\varepsilon_{\infty,\parallel} = 5.95$ according to Ejder [13]. For the phonon frequencies, we use $\omega_{\mathrm{TO,A_1}} = 531\ cm^{-1}$, $\omega_{\mathrm{LO,A_1}} = 733\ cm^{-1}$, $\omega_{\mathrm{TO,E_1}} = 558\ cm^{-1}$, and $\omega_{\mathrm{LO,E_1}} = 740\ cm^{-1}$ according to Deguchi *et al* [11]. As expected, the dispersion curve for the surface phonon–polariton starts from the light line [1–3]. The observed absorption peak appears in the region where the dispersion curve exists.

The dispersion curves of bulk phonon–polaritons having the ordinary (extraordinary) wave's characteristic [14] have been also calculated, though they are not shown herein. Their upper branches are located in the frequency region above $\omega_{\mathrm{A_1,LO}}$ and their lower branches are located in the frequency region below $\omega_{\mathrm{E_1,TO}}$. Therefore, they are not responsible for the observed absorption.

An ATR scan line has been calculated using the wavelength dispersion of the refractive index of KRS-5 given by Haraguchi *et al* [15] and the nominal total-reflection angle of 45°. The calculated ATR scan line is shown by a thick chain line in the left-hand-side panel of figure 1. The intersection of the solid curve and the thick chain line should correspond to the absorption peak position. As can be seen from figure 1, the experimental peak frequency and the frequency read from the intersection show a good agreement. This confirms that the observed absorption peak does indeed correspond to the absorption due to the surface phonon–polariton.

In conclusion, we have succeeded in observing the surface phonon–polariton in wurtzite GaN by the use of the ATR method.

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