Energy transport analysis in a $\text{Ga}_{0.84}\text{In}_{0.16}\text{N}/\text{GaN}$ heterostructure using microscopic Raman images employing simultaneous coaxial irradiation of two lasers

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ABSTRACT

Anisotropic heat transport in a $\text{Ga}_{0.84}\text{In}_{0.16}\text{N}/\text{GaN}$ -heterostructure on a sapphire substrate is observed from microscopic Raman images obtained by utilizing coaxial irradiation of two laser beams, one for heating (325 nm) in the GaInN layer and the other for signal probing (325 nm or 532 nm). The increase in temperatures of the GaInN layer and the underlying GaN layer is probed by the 325-nm and 532-nm lasers, respectively, by analyzing the shift in the Raman peak energy of the higher energy branch of E_2 modes. The result reveals that energy diffuses across a considerable length in the GaInN layer, whereas the energy transport in the perpendicular direction to the GaN layer is blocked in the vicinity of misfit dislocations on the heterointerface. This simultaneous irradiation of two lasers for heat generation and probing is effective in the microscopic analysis of energy transport through heterointerfaces.

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The device operating characteristics of light-emitting devices and transistors deteriorate owing to the heat generation and resultant increase in the temperature of the devices.^{1–3} Furthermore, the population distribution in excitonic states varies with an increase in phonon occupation or phonon temperature,^{4,5} and the change in the distribution reduces the rate of radiation and efficiency.⁵ When the exclusion of heat energy from optical and electronic device design becomes necessary, phonon exclusion from the active layer is required. Various theoretical and experimental studies have been reported on the temperature increase and heat transport in film structures and devices. The effects of crystal defects on thermal energy transport have been extensively discussed in many articles, some of which indicate that the effect of threading dislocations is appreciable when the density is over 10^8 – 10^{10} cm^{-2} .^{6,7} One such article points out that voids are major defects for suppressing thermal transport.⁸

Micro-Raman mapping is a useful tool for the analysis of spatial local temperatures.^{9,10} Heat generation in transistors is an example^{10–12} where the analysis is extended to the temperature gradient

within the crystal.^{12,13} Including this technique, various methods have been studied for the analysis of heat transport across heterointerfaces or superlattices.^{14–22} However, microscopic local imaging of heat transport at a specific defect is still a challenge. In the Raman scattering measurement, two spectra obtained with two laser beams of different wavelengths provide the depth profile of the strain field of a two-layer structure.²³ The analyses of heat transport at a heterointerface or crystal defects require an additional element of simultaneous incidence of two laser beams: the pump (heating) and probe system. In Raman measurements, mode-separated information of phonons can be obtained, which is an advantage compared to the pump-probe method of photo-reflectivity.^{16,24}

In the vicinity of heterointerfaces comprising III-nitride crystals, high strain fields and defects such as misfit dislocations (MDs) or threading dislocations are present in many cases.^{25,26} $\text{Ga}_x\text{In}_{1-x}\text{N}$ alloys are candidate materials for visible color to infrared light emitting devices and solar cells, and they cover a wide lattice parameter range based on the 11% lattice mismatch between InN and GaN.^{27–29} This high

lattice mismatch yields MDs in the vicinity of $\text{Ga}_x\text{In}_{1-x}\text{N}/\text{GaN}$ interfaces and spatial nonuniformity in the alloy composition.^{25–33} Crystal heating at defects is expected to affect carrier dynamics such as nonradiative carrier recombination, which generates further heat energy.^{4,5,34,35} Further, recently, microscopic Raman imaging has been applied to analyses of threading dislocation structures of GaN.³⁶

In this Letter, we will present an analysis of anisotropic heat transport in a heterostructure of $\text{Ga}_{0.84}\text{In}_{0.16}\text{N}/\text{GaN}$ on a sapphire substrate using a microscopic Raman imaging system. We will utilize coaxial two-wavelength laser irradiation, which enables the imaging of the heat transport within the $\text{Ga}_{0.84}\text{In}_{0.16}\text{N}$ film and the underlying GaN layer, and as well as local blockage of heat transport at MDs.

A $\text{Ga}_x\text{In}_{1-x}\text{N}$ (110 nm) film was grown over a GaN film on a sapphire substrate using a metal organic vapor phase epitaxy system.^{30,31} Figure 1(a) shows a reciprocal lattice space mapping of (1105) diffraction. The broad diffraction image of the $\text{Ga}_x\text{In}_{1-x}\text{N}$ layer indicates that the crystal is partially relaxed, while, on another part, pseudomorphic growth occurs on the GaN layer. In this pseudomorphic region, the average GaN mole fraction x was found to be 0.84. No geometrical surface structure was found in the optical microscope image or scanning electron microscope image. The analysis of the p-polarized IR reflectance spectrum for the LO phonon–plasmon coupling mode revealed that the residual electron density of this film was approximately $6 \times 10^{17} \text{ cm}^{-3}$. The higher energy branch of E_2 mode (E_2^H) of the $\text{Ga}_x\text{In}_{1-x}\text{N}$ or GaN film was characterized using a microscopic Raman imaging system using two lasers: 325 nm for the electronic band-to-band excitation and subsequent phonon generation and 325 nm or 532 nm for Raman probing. The measurements were conducted at 295 K. An objective lens with $\text{NA} = 0.5$ and $\times 40$ was utilized. When beams of two different wavelengths were simultaneously incident at the same point on the sample surface, the 532-nm light was focused with a diameter of approximately $0.65 \mu\text{m}$, while the 325-nm light was diffused at an approximate diameter of $7 \mu\text{m}$. Under this condition, the local increase in temperature by the heat generation was probed. The relaxation energy of excited carriers to the band bottoms

FIG. 1. Reciprocal lattice space mapping of x-ray diffraction. The respective notations of q_c and q_m are expressed as $5/c$ and $2/a$. Here, c and a are the lattice parameters of the c and a axes, respectively.

is expected to be 27% of the excitation energy. Further energy transfer to the lattice vibration system is possible through nonradiative carrier recombination. The thermal properties were analyzed by the two-dimensional mapping of the E_2^H energy probed by the Raman signal using the 325-nm or 532-nm light. The energy shift of E_2^H of $\text{Ga}_x\text{In}_{1-x}\text{N}$ was obtained by varying the power of the 325-nm laser light focused on the sample surface. The density of free carriers generated by a 1 mW excitation of the focused 325-nm beam was estimated to be approximately $5 \times 10^{18} \text{ cm}^{-3}$ or less for the excitation laser power of 1 mW, which is based on the photoluminescence (PL) lifetime less than 500 ps for the band–band transition. Carrier diffusion in the $\text{Ga}_x\text{In}_{1-x}\text{N}$ layer further decreases the carrier density to approximately one quarter. In simultaneous excitation, the carrier density is estimated to be less than 10^{17} cm^{-3} for the excitation laser power of 10 mW. In these cases, the thermal conduction by free carriers can be neglected.⁷

Figure 2(a) shows the PL intensity mapping in a $45\text{-}\mu\text{m}$ square region, and spectrum examples are shown in Fig. 2(b). These dark lines are parallel to the directions of $\langle 1\bar{1}00 \rangle$ and intersect mutually at an angle of 60° . These types of dark lines and areas are attributed to nonradiative recombination (NR) in the vicinity of MDs at the $\text{Ga}_{0.84}\text{In}_{0.16}\text{N}/\text{GaN}$ interface.^{25,26} Figure 2(c) shows a magnified image of the square area marked by a solid line in Fig. 2(a). A bright triangular region is surrounded by a dark triangular rim. Some dark lines, for example, line A in Fig. 2(c), are narrow, which suggests that the carrier diffusion length is approximately $0.5 \mu\text{m}$ or less. This value is in the range of previously reported values of $0.1\text{--}3 \mu\text{m}$ for the alloy composition x of $0.70\text{--}0.97$.^{32,33,37,38} Small spikes, for example, at 2.71 eV , originate from the mismatch of measurement conditions of PL and lamp light for sensitivity calibration using an interference filter in our system. It is found that PL spectra in the bright region have single peak with its maximum at approximately 2.825 eV and that PL spectra in the dark region have another peak superposed at 2.775 eV . Figure 2(d)

FIG. 2. Imaging of PL properties, intensity mapping for a $45\text{-}\mu\text{m}$ square region (a), examples of PL spectra in bright and dark regions (b), intensity mapping in a magnified $8\text{-}\mu\text{m}$ square region of the solid-line square (c), and PL intensity ratio of the peaks at 2.775 eV and 2.825 eV (d).

shows a mapping image of the PL intensity ratio of the peak at 2.775 eV and that at 2.825 eV. It appears that this figure reflects the intensity mapping structure, while it presents further information. The higher energy side peak grows as the total PL intensity increases. When we compare PL spectra at positions B and C, both of which are dark regions, the second peak is found to be strong at position C, while at position B, it is weak. This feature suggests that the crystal quality is recovering at position B even though the PL intensity is still low. It is believed that regions with lower peak energy have a higher InN mole fraction than those with higher peak energy. Further, crystal quality is improved with an increase in the film thickness.

Figure 3(a) shows the mapping images of the Raman peak energy using 325-nm laser light with a power of 1.1 mW. A specimen of the obtained spectra is shown in Fig. 3(d). The observed region is on the inside of the white broken square line in Fig. 2(a). As this laser light is absorbed within the depth of approximately 100 nm from the surface, the Raman peak is attributed to E_2^H of the $\text{Ga}_x\text{In}_{1-x}\text{N}$ layer. The image of the E_2^H peak energy shows a geometric structure similar to that of the PL peak intensity. Figure 3(b) shows the mapping image of the peak energy obtained with a higher laser intensity of 11 mW, where the triangular structure can be clearly observed. The difference in the peak energy between these two laser intensities is plotted in Fig. 3(c), where any triangular structure cannot be observed. Figure 3(e) shows the histogram plotted for peak energy distribution. The mean peak energy shift with increased laser power is approximately 0.5 cm^{-1} .

The parameters to analyze the mode energies are listed in Table I.^{39–46} Here, we assume that the shift of mode energy $\Delta\omega$ by strain is expressed as $2a\varepsilon_{xx} + b\varepsilon_{zz}$, which is transformed to

$$\Delta\omega = (2a - 2bv/(1 - \nu))\varepsilon_{xx}. \quad (1)$$

FIG. 3. Raman spectrum mapping images for the 325-nm laser, (a) E_2^H peak energy for 1.1-mW excitation, (b) E_2^H peak energy for 11-mW excitation, (c) energy shift of E_2^H of the $\text{Ga}_x\text{In}_{1-x}\text{N}$ layer by increasing the excitation power, (d) Raman spectrum example, and (e) histogram of the distribution in the peak energy decrement by increasing the excitation power.

Here, ε_{xx} , ε_{zz} , and ν are the strain in the x direction, the strain in the z direction, and Poisson ratio, respectively. The notations of a and b are the deformation potentials. When we adopt the potential values of $a = 513 \text{ cm}^{-1}$ and $b = 597 \text{ cm}^{-1}$ for E_2^H mode, $\nu = 0.25$ for GaN, and the mode energies for $\text{Ga}_x\text{In}_{1-x}\text{N}$ alloys as reported in Ref. 47, the obtained observed mode energy is consistent with the GaN mole fraction and strain values obtained by the XRD analysis for the pseudomorphic growth of the $\text{Ga}_{0.84}\text{In}_{0.16}\text{N}$ layer on the GaN layer in the triangle structure. Here, the parameters of $\text{Ga}_x\text{In}_{1-x}\text{N}$ excluding mode energies were obtained by the linear interpolation of the values of GaN and InN.

When we assume that the temperature increase of the sapphire substrate excluding the vicinity of the interface is negligible, and the Poisson ratio is constant, the energy shift of the E_2^H peak by increasing the laser power of 325 nm, $\Delta E(E_2^H)$, is expressed as follows by considering the thermal strain induced by heating:

$$\Delta E(E_2^H) \approx \Delta E(E_2^H : T - T_0, 0) + C_E \cdot l_{th}/l \cdot (T - T_0). \quad (2)$$

Here, C_E is the coefficient presenting the dependence of mode energy on in-plane strain ε_{xx} shown in Eq. (1). $\Delta E(E_2^H : T - T_0, 0)$ is the decrease in mode energy with an increase in temperature from T_0 to T for the bulk crystal without a gradient in temperature, i.e., no thermal stain. The temperature coefficient of mode energy for unstrained $\text{Ga}_x\text{In}_{1-x}\text{N}$ was obtained by linear interpolation between the values of free standing GaN and InN. Using the parameters in Table I, $\Delta E(E_2^H)$ is found to be $1.6 \times 10^{-2} \cdot \Delta T \text{ (cm}^{-1}\text{)}$. The energy shift of 0.5 cm^{-1} for E_2^H suggests an increase of 31 K with an increase in the excitation power of the 325-nm light. The featureless image of the decrement of Raman peak energy shown in Fig. 3(c) indicates that thermal energy transport is independent of the defects on the $\text{Ga}_x\text{In}_{1-x}\text{N}$ /GaN interface. Two mechanisms are considered: the long diffusion length of thermal energy with respect to the size of the triangle and dominant thermal conduction on the surface side $\text{Ga}_x\text{In}_{1-x}\text{N}$ layer with higher crystal quality than that of the crystal in the vicinity of MDs.

Figure 4(a) shows an example of the Raman spectra obtained by the 532-nm line with and without the simultaneous irradiation of the 325-nm light. The spectrum peak at approximately 569 cm^{-1} is attributed to E_2^H of the GaN film. Figure 4(b) shows the mapping image of the peak energy obtained by Lorentzian fitting for the region indicated by the white solid square line in Fig. 2(a). The variation in the mode energy in this area is within 1.1 cm^{-1} , which corresponds to the strain variation of approximately 0.12%. When the 325-nm light with a power of 20 mW was incident on the sample, mode energies in some regions were found to shift to the lower energy side, which indicates an increase in temperature. A pair of sample spectra is presented in Fig. 4(a). The obtained mapping of the decrement in mode energy is plotted in Fig. 4(c). The observed image exhibits a structure similar to the triangle observed by the PL measurement shown in Fig. 2. The decrease in E_2^H energy of the GaN film in the dark PL region including MDs is lower than that of the region exhibiting brighter PL emission. This result indicates that the temperature increase in the GaN layer within the triangle exhibiting bright PL emission of the $\text{Ga}_x\text{In}_{1-x}\text{N}$ layer is higher than that of the region outside the triangle rim.

Most of the 325-nm laser is absorbed inside the GaInN layer, where heating energy is generated, while the 532-nm laser penetrates the GaInN layer to the GaN layer. Thus, the Raman signal using the 532-nm laser is dominated by the signal from the GaN layer. Thus,

TABLE I. Parameters for anisotropic strain and lattice vibration of GaN and InN.

	a (cm ⁻¹)	b (cm ⁻¹)	ν	l_{th} (10 ⁻⁶ K ⁻¹)	$d\omega/dT$ (10 ⁻² cm ⁻¹ K ⁻¹)	ω_0 (cm ⁻¹)
GaN E ₂ ^H	513 ³⁹	597 ³⁹	0.183 ⁴⁰ 0.25–0.33 ⁴¹	4.2 ⁴⁴	0.87 ⁴⁰	568.5 ⁴¹ , 567.6 ⁴⁶
A ₁ ^{LO}	664 ⁴¹	882 ⁴¹			2.4 ⁴¹	736.4 ⁴¹
InN E ₂ ^H	938 ⁴²	407 ⁴²	0.21 ⁴² 0.27 ⁴⁰	3.83 ⁴³	13 ⁴⁵	494.6 ⁴²
A ₁ ^{LO}	901 ⁴²	587 ⁴²			6.0 ⁴⁵	582.3 ⁴²

when using the 325-nm laser as the probe laser, the temperature increase in the GaInN layer is probed, and when using the 532-nm laser as the probe laser, the temperature increase in the GaN layer by the heat transport from the GaInN layer to the GaN one is observed.

Therefore, it is believed that the extent of MDs blocks the heat transport to the GaN layer. These experimental results indicate that the lateral heat transport in the Ga_xIn_{1-x}N layer takes place primarily via the crystal in the surface side region with improved crystal quality, while the transport to the underlying GaN layer is mainly blocked in the vicinity of MDs. The schematic of this model is shown in Fig. 4(d). The increase in temperature within the triangle is estimated to be approximately 20 K, which is an average value in the region probed by the 532-nm light.

In conclusion, the pump and probe method in Raman spectroscopy is valid for analyzing energy or heat transport across the hetero-interface. The pump laser light generates heat energy by carrier energy relaxation in the bands, and the Raman signal is obtained by the probe laser light. In this article, we have selectively observed the Raman signal of the GaN layer by using the 532-nm laser, which allows the

separation of the excitation position (GaInN layer) and probing position (GaN layer). It is observed that the heat transport across the hetero-interface is blocked in the vicinity of MDs. It is considered that this method is applicable to the analyses of single threading dislocation at heterointerfaces when stable measurements with spectrum fitting resolution higher than 0.1 cm⁻¹ are available. It is expected to be possible to discuss the dependence of heat conduction on the structure of the dislocation core.

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FIG. 4. Variation in E₂^H mode energy of the GaN layer by 325-nm laser irradiation, which is probed by the 532-nm laser, (a) spectra with and without irradiation of the 325-nm laser, (b) image of E₂^H peak energy obtained by the 532-nm laser without irradiation of the 325-nm laser, (c) decrease in the E₂^H peak energy by the irradiation of 20 mW of the 325-nm laser, and (d) schematic of the heat transport for two regions with and without MDs beneath the laser spot. The solid curves in (a) are the best-fit Lorentzian functions.

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