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

**ABSTRACT**

Anisotropic heat transport in a Ga$_{0.84}$In$_{0.16}$N/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.

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. Furthermore, the population distribution in excitonic states varies with an increase in phonon occupation or phonon temperature, and the change in the distribution reduces the rate of radiation and efficiency. 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$^{-2}$. One such article points out that voids are major defects for suppressing thermal transport.

Micro-Raman mapping is a useful tool for the analysis of spatial local temperatures. Heat generation in transistors is an example where the analysis is extended to the temperature gradient within the crystal. Including this technique, various methods have been studied for the analysis of heat transport across heterointerfaces or superlattices. 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. 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.

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

In this Letter, we will present an analysis of anisotropic heat transport in a heterostructure of Ga\textsubscript{0.84}In\textsubscript{0.16}N/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 Ga\textsubscript{0.84}In\textsubscript{0.16}N film and the underlying GaN layer, and as well as local blockage of heat transport at MDs.

A Ga\textsubscript{In}\textsubscript{N} (110 nm) film was grown over a GaN film on a sapphire substrate using a metal organic vapor phase epitaxy system.\textsuperscript{30,31} Figure 1(a) shows a reciprocal lattice space mapping of (1T05) diffraction. The broad diffraction image of the Ga\textsubscript{In}\textsubscript{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 \( 6 \times 10^{17} \) cm\(^{-3}\). The higher energy branch of E\textsubscript{2} mode (E\textsubscript{21}) of the Ga\textsubscript{In}\textsubscript{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 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 \( \mu \)m, while the 325-nm light was diffused at an approximate diameter of 7 \( \mu \)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 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\textsubscript{21} energy probed by the Raman signal using the 325-nm or 532-nm light. The energy shift of E\textsubscript{21} of Ga\textsubscript{In}\textsubscript{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 \( 5 \times 10^{16} \) 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 Ga\textsubscript{In}\textsubscript{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.

Figure 2(a) shows the PL intensity mapping in a 45-\( \mu \)m square region, and spectrum examples are shown in Fig. 2(b). These dark lines are parallel to the directions of (1T00) 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 Ga\textsubscript{In}\textsubscript{N}/GaN interface.\textsuperscript{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 \( \mu \)m or less. This value is in the range of previously reported values of 0.1–3 \( \mu \)m for the alloy composition x = 0.70–0.97.\textsuperscript{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 (d).

![FIG. 1.](image1.jpg) Reciprocal lattice space mapping of x-ray diffraction. The respective notations of \( q_c \) and \( q_m \) are expressed as 5/1 and 2/1. Here, c and a are the lattice parameters of the c and a axes, respectively.

![FIG. 2.](image2.jpg) Imaging of PL properties, intensity mapping for a 45-\( \mu \)m square region (a), examples of PL spectra in bright and dark regions (b), intensity mapping in a magnified 8-\( \mu \)m square region of the solid-line square (c), and PL intensity ratio of the peaks at 2.777 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.

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 $2a_{\text{xx}} \chi + b_{\text{xx}}$, which is transformed to

$$\Delta \omega = (2a - 2b\nu/(1 - \nu))e_{\text{xx}}. \quad (1)$$

The energy shift of 0.5 cm$^{-1}$ in the peak energy between these two laser intensities is plotted in Fig. 4(c). The observed image exhibits a structure similar to that of the PL peak intensity. Figure 3(b) shows the mapping image of the peak at 2.825 eV and that at 2.825 eV. It appears that this figure reflects the thermal conduction on the surface side GaN, thermal energy with respect to the size of the triangle and dominant strain is expressed as $2\chi_{\text{xx}}$.

The notations of GaN, InN, and Ga$_{0.84}$In$_{0.16}$N alloys as reported in Ref.47, the temperature increase in the GaN layer $\Delta E_{\text{H}}(T)$ is expressed as follows by considering the thermal strain induced by heating

$$\Delta E_{\text{H}}(T) \approx \Delta E_{\text{H}}(T = T_0, 0) + C_{\text{E}} \cdot l_{\text{b}/(1 - \nu)} \cdot (T - T_0). \quad (2)$$

The temperature coefficient of mode energy on in-plane strain $e_{\text{xx}}$ shown in Eq. (1). $\Delta E_{\text{H}}(T = T_0, 0)$ 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 Ga$_{0.84}$In$_{0.16}$N was obtained by linear interpolation between the values of free standing GaN and InN. Using the parameters in Table I, $\Delta E_{\text{H}}(T)$ is found to be $1.6 \times 10^{-2}$ - $\Delta T$ (cm$^{-1}$). The energy shift of 0.5 cm$^{-1}$ for $E_{\text{H}}$ 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 Ga$_{0.84}$In$_{0.16}$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 Ga$_{0.84}$In$_{0.16}$N layer with higher crystal quality than that of the crystal in the vicinity of MDs.

FIG. 3. Raman spectrum mapping images for the 325-nm laser, (a) $E_{\text{H}}$ peak energy for 1.1-mW excitation, (b) $E_{\text{H}}$ peak energy for 11-mW excitation, (c) energy shift of $E_{\text{H}}$ of the Ga$_{0.84}$In$_{0.16}$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.
when using the 325-nm laser as the probe laser, the temperature increase in the GaN 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 GaInN 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 heterointerface. 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 heterointerface 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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