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A Novel Method to Measure Absolute Internal Quantum Efficiency in III-Nitride Semiconductors by Simultaneous Photo-Acoustic and Photoluminescence Spectroscopy*

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SUMMARY Internal quantum efficiency (IQE) is usually estimated from temperature dependence of photoluminescence (PL) intensity by assuming that the IQE at cryogenic temperature is unity. III-nitride samples, however, usually have large defect density, and the assumption is not necessarily valid. In 2016, we proposed a new method to estimate accurate IQE values by simultaneous PL and photo-acoustic (PA) measurements, and demonstratively evaluated the IQE values for various GaN samples. In this study, we have applied the method to InGaN quantum-well active layers and have estimated the IQE values and their excitation carrier-density dependence in the layers.

key words: *internal quantum efficiency, recombination, PAS, PL*

1. Introduction

III-nitride-based optical devices have been wide-spreading for their advantages of high efficiency and device lifetime, and further improvement in light-output efficiency has been expected for global energy saving. The external quantum efficiency (EQE) in these devices is described by the product of internal quantum efficiency (IQE), current injection efficiency (CIE) and light extraction efficiency (LEE), among which, the IQE is an important factor reflecting the crystal quality of the active layers. The IQE values in III-nitride materials are usually estimated from temperature dependence of photoluminescence (PL) intensity [1]–[4]. In this method, the IQE at cryogenic temperature (below 10 K) is assumed as unity, and the IQE at room temperature (RT) is estimated as the ratio of the PL intensity at RT to that at low temperature. The assumption means that nonradiative recombination processes at extremely low temperature are completely suppressed. III-nitride materials, however, are usually highly defective and the nonradiative processes can be still active even at low temperature [5]. Thus, the conventional method could not necessarily give accurate IQE values, and more accurate methods are needed. Recently, Usami *et. al*. proposed a method to estimated absolute IQE values of active layers in LEDs by combination of photocurrent and PL measurements [6]. Kojima *et. al*. proposed another method in which they accurately measure absolute PL intensity using an integrating sphere [7]. Nonradiative recombination flow is, however, not directly observed, but indirectly computed in their methods. In our previous study, we preliminary proposed a new experimental method to estimate the absolute IQE values, in which both the radiative and nonradiative recombination flows are directly observed by simultaneous photo-acoustic (PA) and PL measurements. We also demonstratively performed the measurements in GaN films with different qualities [8], [9]. In this method, the PA and PL measurements are simultaneously carried out for the same samples at RT, and IQE values (at RT) can be accurately estimated only from the RT measurements without cooling samples. Generally, photo-excited carriers recombine either radiatively or nonradiatively, after the photo-excitation, and photons and phonons are generated by the radiative and nonradiative processes, respectively. The PL measurement detects “light” generated by the radiative recombination and the PA measurement detects “heat” generated by the nonradiative recombination [10]. Thus, the PA/PL simultaneous measurements can acquire information on both the radiative and nonradiative recombination processes, and this could makes it possible to measure the IQE values more accurately compared with the conventional method in which only PL signals are detected and the doubtful assumption mentioned above is needed.

In this work, we have performed the simultaneous PA and PL measurement for an InGaN quantum-well (QW) structure which is usually used as an active layer for a III-nitride based optical device, and have successfully estimated its absolute IQE value. The results are compared with those of the conventional method, and it is found that the results are significantly different. This suggests that the assumption in the conventional method is not correct in the InGaN-QW sample.

2. Experimental

The sample used in this work was an InGaN-QW film grown by metal-organic chemical vapor deposition (MOCVD).

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Figure 1 shows the experimental set up for the simultaneous PA and PL measurement. All the measurements were performed at RT. In the measurement, a semiconductor laser diode was used as an excitation source. The excitation wavelength was 405 nm, and the excitation light is absorbed only in the QW layer (transparent in barrier layers). The laser light was modulated with 80 Hz by a chopper and focused on the surface of the sample in a closed cell through a quartz window. Spot size was around 100μm. Light emission (PL) from the sample was collected by a lens into a spectrometer (Ocean Optics, USB4000). On the other hand, lattice vibration (phonon) generated by nonradiative recombination (heat generation) was detected as a sound signal, which is called a PA signal, by a high-sensitive microphone, and the PA signal with the chopper frequency was amplified by a lock-in amplifier [11].

Next, we will describe how to obtain the IQE values from the PA/PL simultaneous measurements. Although PL and PA signals are considered to be proportional to radiative and nonradiative recombination rate, respectively, the intensity of the two signals cannot be compared directly, and thus the IQE values also cannot be derived. The signals, however, can be compared by utilizing nonlinear behavior in excitation power dependence. PL and PA intensities usually show superlinear or sublinear dependence with respect to the laser excitation power. In the measurements, the energy supplied to the sample by laser light excitation is emitted either in the form of "light" or "heat" by radiative or nonradiative recombination processes, respectively. Therefore, in case PL intensity shows a superlinear behavior, PA intensity should show a sublinear behavior. Conversely, in case PL intensity shows a sublinear behavior, PA intensity should show a superlinear behavior. This complementary relationship enables the comparison of the two signals' intensities to obtain the IQE values. Since the total energy of "light" and "heat" generated in the sample should be proportional to the laser excitation power, the amounts of the upward and downward shifts from linear relationship in PA and PL intensities should be cancelled in total (PA+PL) signal intensity. Therefore, the comparison becomes possible when the PA or PL signal is rescaled so that the sum of the two signals can show linear dependence with respect to excitation power. Then, the ratio between the "light" and "heat" energy generated in the sample can be obtained from the rescaled PA and PL signals, and the IQE value is determined for each laser excitation power.

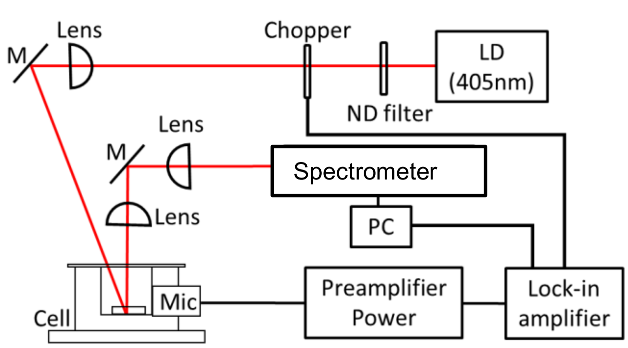

Fig. 1 Experimental set up for the simultaneous PA and PL measurements.

3. Results and Discussions

First, we measured the excitation wavelength dependence of PA signal intensity in order to confirm that the PA measurement detects the "heat" generated by nonradiative recombination process of photo-excited carriers. In the measurement, Xe-lamp light is monochromatized by a monochrometer and used for the excitation. Figure 2 shows the excitation wavelength dependence of PA signal intensity for the InGaN-QW sample. The PL and photoluminescence excitation (PLE) spectra for the same sample are also plotted in the figure. The PL peak appears at ~485 nm, and the both PA and PLE signals increase with decreasing the excitation wavelength. Since the PLE spectrum is considered as similar as absorption spectrum in the InGaN-QW layer, the similarity between the PA and PLE spectra shows that the PA signal corresponds to the heat generation by nonradiative recombination of photo-excited carriers in the InGaN-QW layer, although the origin of peak structure at around 440nm is unclear.

Next, we measured the excitation power dependence of the PA intensity and the PL integrated intensity for the InGaN-QW emission. The results are shown in Fig. 3(a). It is shown, in this figure, that the PA signal intensity slightly shows sublinear dependence with respect to the excitation power while the PL signal shows superlinear dependence. These nonlinear behaviors are shown more clearly in Fig. 3(b), in which the ratio of PA (PL) signal intensity to the laser excitation power is plotted as a function of the excitation power. As mentioned above, the sum of the PA and PL energies should be proportional to the excitation power because the energy supplied to the sample by laser light excitation is emitted either in the form of "light" or "heat" by

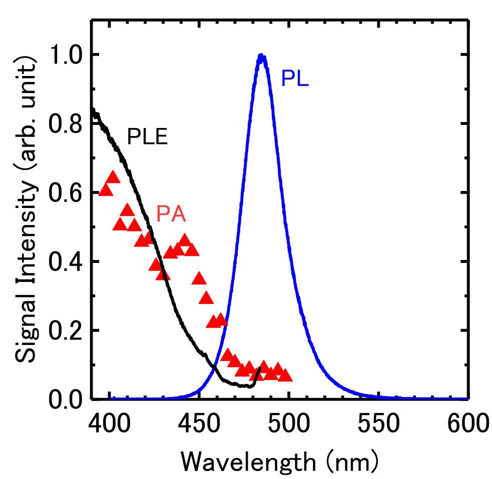

Fig. 2 Excitation wavelength dependence of PA signal (closed triangle) for an InGaN-QW sample. The PL (blue line) and PLE (black line) spectra for the same sample are also plotted.

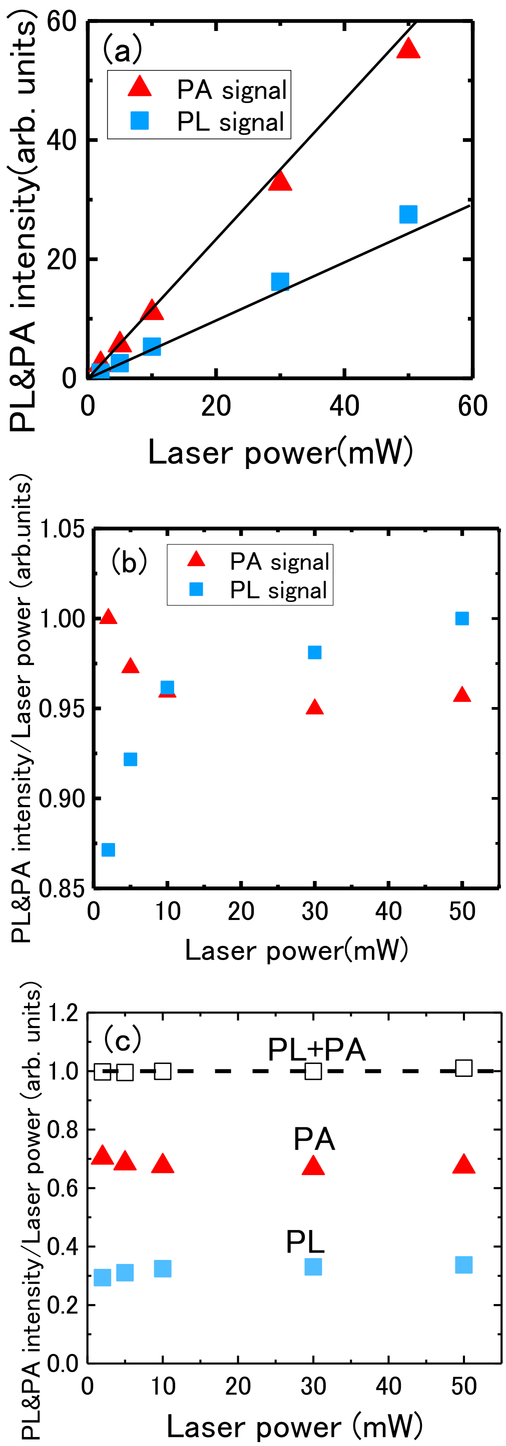

Fig. 3 (a) Excitation power dependence of PL (closed square) and PA (closed triangle) signal intensities. (b) Ratio of PL and PA signal intensities to the excitation laser power as a function of the excitation power. (c) The rescaled graph so that the sum of the two ratios (PL+PA) can be constant. The sum (PL+PA) is also shown in the figure.

radiative or non-radiative processes, respectively. Thus, PA and PL signals can be compared by rescaling the vertical axis in Fig. 3(b) so that the sum of the two signals can show linear dependence with respect to the excitation power. Such

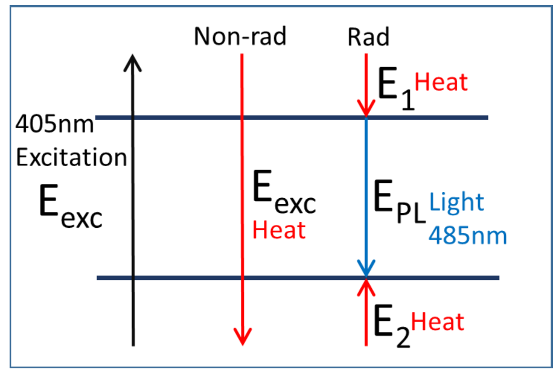

Fig. 4 Two processes of photo-excited carriers are schematically shown. The radiative process also generates "heat" in intra-band relaxation.

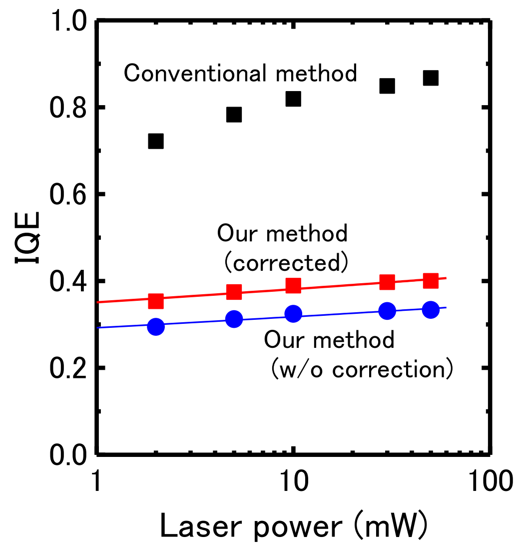

Fig. 5 Estimated absolute IQE values by our method (closed circle) and conventional method (closed square) as a function of excitation laser power. The estimated values without consideration of the intra band energy relaxation effect, are also plotted.

a rescaled graph is shown in Fig. 3(c), where the sum of the two ratios (PL+PA) is almost constant. Thus, IQE values can be estimated by using Fig. 3(c).

It should be noted that the IQE values thus estimated should be corrected by consideration of the "intra-band relaxation effect". Since the excitation photon energy is somewhat larger than the emission photon energy in this measurement, photo-excited carriers have some excess energy just after photo-excitation. The excess energy is emitted in the form of phonons inside the bands, and heat generation takes place even in radiative recombination paths by the intra-band energy relaxation and the PA measurement undesirably detects this heat generation as shown in Fig. 4 schematically. The correction for this effect can be made by just multiplying a factor of E_{exc}/E_{PL}, where E_{exc} and E_{PL} are the photon energies of excitaion and photoluminescence, respectively. Figure 5 shows the IQE values estimated by this method from the data in Figs. 3(a)–(c) as a function of laser excitation power. The IQE values without consideration of the intra-band energy relaxation effect, are also plotted in this figure. It is found, in Fig. 5, that the IQE value is around 40 % for the InGaN-QW sample at RT, and that the IQE gradually increases with excitation power. The power dependence is probably due to the filling-up of non-radiative centers by photo-excited carriers.

Finally, the estimated IQE values were compared with that estimated by the conventional method (temperature dependence of PL intensity). The temperature dependent PL measurements were performed at several excitation laser power conditions using the same light source for the same samples. The excitation power dependence of the estimated IQE values for the two methods are also plotted in Fig. 5. It is shown that the estimated IQE values are significantly different between our method and the conventional method. For instance, the IQE value at RT is estimated as 40 % for 50 mW excitation by our method while the conventional method gives the IQE value of 90 %. These results indicate that nonradiative recombination processes are still active even at extremely low temperature in the InGaN-QW sample and that the conventional IQE estimation method cannot necessarily give accurate values.

4. Conclusions

We have proposed a novel method to measure accurate absolute IQE values by simultaneous PA and PL spectroscopy in III-nitride semiconductors, in which “light” and “heat” generation in radiative and non-radiative recombination processes, are directly detected by PA and PL measurements, respectively, and the IQE values can be accurately estimated from correlative relationship between the two signals. The measurements were demonstratively performed for a blue-emitting InGaN-QW sample, and the estimated IQE values were compared with those estimated from the conventional method. It is found that the conventional method cannot necessarily give accurate IQE values and that the proposed method is a promising way for accurate IQE estimation.

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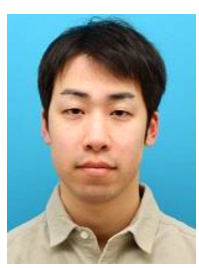

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