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Two-photon absorption and multiphoton-induced photoluminescence of bulk GaN excited below the middle of the band gap

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Optical nonlinearity in the yellow luminescence (YL) band of GaN was investigated using thick bulk samples. Transient pump–probe measurements revealed strong transmission changes due to two-photon absorption (TPA) even at the middle of the YL band. The TPA coefficient evaluated reaches ~5 cm/GW at about 1.3 eV, which was as large as the mid-gap resonance. The TPA spectrum clearly showed that the observed large nonlinearity originated from the YL band. On the basis of efficient TPA in the YL band, relaxation processes in the multiphoton-induced photoluminescence excitation spectrum were also investigated. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1587260]

The development of fabrication technologies for GaN and III nitrides has resulted in numerous device applications including blue lasers.[1] Recently, the optical nonlinearities of these materials have attracted increased interest as factors for determining device performance.[2–11] In addition to the technological importance, these nonlinear absorption studies also provide important insights into the fundamental physics of materials, such as band structures,[6] relaxation processes,[7–10] and electron–phonon interactions.[11] One characteristic of GaN observed in photoluminescence (PL) spectra is yellow luminescence (YL).[12–15] Kim *et al.* reported multiphoton-induced PL in the YL band and found that the multiphoton PL excitation (PLE) spectrum exhibits resonance that is attributable to the mid-gap defect states.[6] Sun *et al.* obtained a large two-photon absorption (TPA) coefficient below the band gap and demonstrated two-photon-induced YL imaging.[3] However, the nonlinear optical properties in the YL band including the TPA coefficients remain largely unclear. This letter presents experimental results for an efficient TPA process associated with the YL band.

Although the multiphoton-induced YL excitation spectrum exhibits only small signals below the middle of the band gap ($E_g/2$), the power dependence clearly reveals a two-photon contribution. Furthermore, TPA coefficients as large as that near the band tail are apparent in the transient pump–probe spectrum in the middle of the YL band. The TPA-related relaxation processes are also discussed by comparing PLE and TPA spectra.

We used a freestanding bulk GaN sample with a thickness of ~420 μm. The absence of a substrate allowed precise evaluation of the nonlinear response. The electron concentration in the sample was estimated to be $\sim 10^{19}$ cm^{-3}. A mode-locked Ti:sapphire laser (pulse width ~100 fs, repetition rate 76 MHz, peak power density ~100 GW/cm^2) was employed as a high-energy, nonresonant excitation light source for nonlinear optical measurements. The tunable wavelength of the laser was 1.25–1.77 eV, and it ranges from $E_g/2$ to the middle of the YL band. The PL signal was dispersed by a 50 cm monochromator with interference filters and detected by a charge-coupled device (CCD). In transient time measurements, linearly polarized pump–probe pulses (intensity ratios of 95:5, respectively; parallel polarization) were focused onto the same point inside the sample. The pump was chopped, and the change in transmission was measured using a lock-in amplifier. All measurements were performed at room temperature.

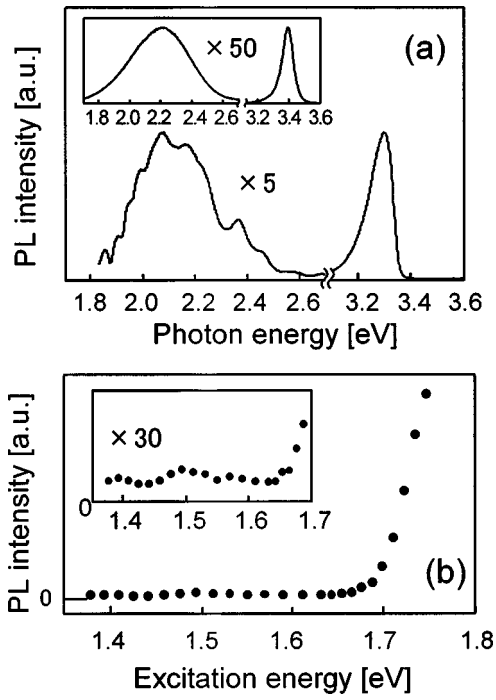

FIG. 1. (a) Multiphoton PL spectrum excited with 1.5 eV photons. Inset: PL excited by a He–Cd laser. The YL ranges from 1.8 to 2.6 eV. (b) Multiphoton PLE spectrum detected at 2.4 eV, corresponding to the higher energy side of the YL. Inset: Expanded view below $E_g/2$.

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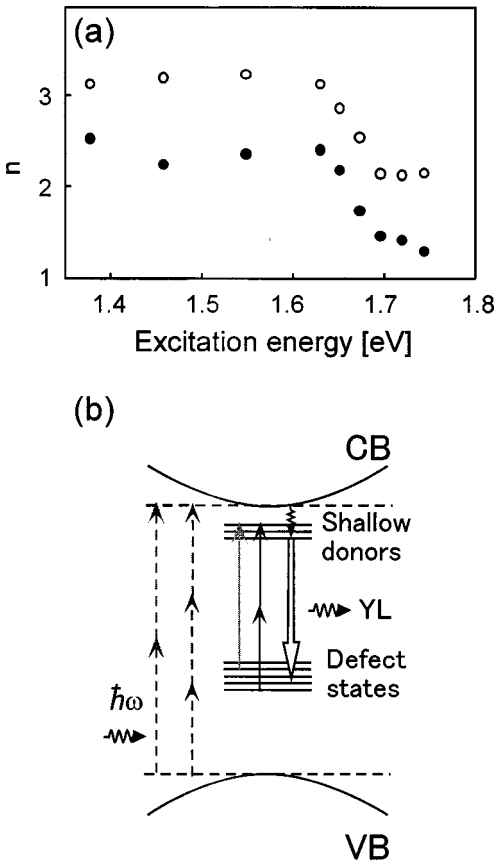

FIG. 2. (a) Excitation energy dependence of multiphoton PL. Closed and open circles correspond to YL and UV luminescence, respectively. (b) Schematic of the carrier transition processes observed in the YL band. Solid and gray arrows indicate excitation energy $<E_g/2$ and $>E_g/2$, respectively.

Figure 1(a) shows a typical multiphoton PL spectrum. The use of 1.5 eV excitation energy allows the possibility of both a YL band transition by two photons as well as an interband transition by three photons. The PL spectrum for excitations above E_g is shown in the inset for comparison. Since the PL peaks in both spectra are identical, the peak at 3.4 eV can be attributed to the band edge, and the 2.2 eV peak can be assigned to YL.

The ultraviolet (UV) luminescence is about two orders of magnitude greater than YL, suggesting that the sample has a standard doping level.[15] In contrast to the sharp band-edge emission, the YL is a broad resonance. This indicates that the YL structure is characterized by a number of deep levels with different energies.[7]

Figure 1(b) shows a typical excitation energy dependence of YL intensity, i.e., the multiphoton PLE of a YL band. Note that the YL intensity at other detection energies exhibited similar behavior. The sharp PLE increase at $E_g/2$ is attributed to two-photon resonance in the band gap. Below $E_g/2$, YL persists but at a much lower intensity than that of the band-gap excitation. In addition, as shown in the inset in Fig. 1(b), the PLE in this region produces a less-structured signal. These results are not due to an inefficient TPA in the YL band and are discussed later. First, it is necessary to consider the absorption and relaxation processes observed in the PLE.

The YL dependence on the excitation intensity was investigated to clarify how many photons contributed to PLE. Figure 2(a) is a plot of the fitting values (n) at different excitation energies based on a power law fit of the excitation intensity (I^n). In Fig. 2, the closed circles denote n in the YL. For comparison, the excitation intensity dependences of UV luminescence were also plotted (open circles). These two luminescences exhibit different magnitudes over the entire range of excitation energies examined. Near the band-gap resonance in the PLE, the observed intensity dependence of the YL is characterized by $n=1.5$, indicating that two processes contribute to the signal. One process is two-photon resonance in the band gap [indicated by dashed gray arrows in Fig. 2(b)]. In this process, the excited carriers can relax to the YL band and then emit luminescence. The second process is a direct transition within the YL band [shown by gray arrows in Fig. 2(b)]. The broad structure of the YL plot suggests that optically excited carriers can be activated by thermal phonons. Therefore, one-photon excitation could contribute to the total YL signal despite excitation with photons below the detection energy. Below 1.65 eV (i.e., $\ll E_g/2$), the YL again appears to be generated by a combination of two- and three-photon processes, while the UV luminescence has a fitting value n of ~ 3, which indicates a pure three-photon transition [shown by dashed black arrows in Fig. 2(b)]. This is reasonable because the high excitation energy of our focusing beam allows the possibility of this interband transition to be excited by three-photon absorption. Since the UV luminescence is due solely to a three-photon transition, TPA from the defect states to the conduction band edge cannot contribute to the YL. Moreover, the crossover in YL remains far below $E_g/2$. These results thus indicate that the two-photon contribution originates from the TPA directly through the YL band in the manner shown by the solid black arrows in Fig. 2(b).

Transient time measurements were employed for direct investigation of TPA in the YL band. Figure 3(a) shows typical transmission changes as a function of the delay time between pump and probe pulses. Dramatic changes in transmission were observed despite the excitation being far below $E_g/2$. The signal profile in the vicinity of zero delay is consistent with that of the autocorrelation of the incident pulse. Therefore, the observed transmission change only reflects the instantaneous electronic response, suggesting the absence of a linear transition, such as a transition from the valence band to a deep level. In addition, the transmission change is found to be linearly dependent on the pump power. This behavior indicates that the signal consists of a TPA process that involves one pump photon and one probe photon. The TPA coefficient was found to be 0.2 cm/GW at 1.5 eV. We obtained this value from the excitation power density of 100 GW/cm^2 and the estimated interaction length of 2.5 μm where the pump and probe pulses overlap. It is important to note that an additional asymmetrical offset signal can be observed at higher excitation intensities. This offset is proportional to I^2, suggesting saturated transparency in the YL band. Note that similar autocorrelation TPA behavior has been observed only for excitation near the bandgap.[3] The use of thick bulk GaN in this study allows intense TPA signals to be observed even below $E_g/2$.

The observed linear power dependence indicates that the transmission change excludes the three-photon contribution

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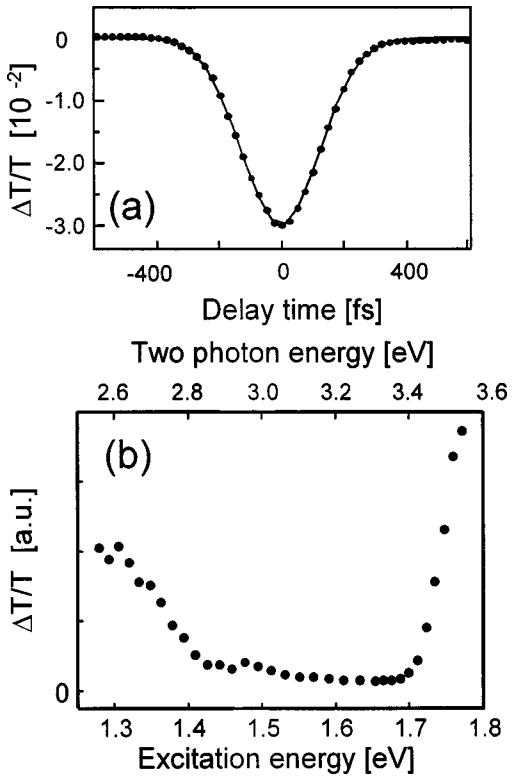

FIG. 3. (a) Decrease in TPA-induced transmission at 1.5 eV excitation. The solid line indicates a Gaussian fit. (b) TPA spectrum obtained from changes in transmission at various excitation energies (E_{ex}). The upper axis corresponds to $2\times E_{ex}$.

observed in Fig. 2(a). Furthermore, it is possible to directly evaluate the TPA spectrum in the YL band by changing the excitation energy. Figure 3(b) shows such a TPA spectrum. Above 1.7 eV, a sudden increase is observed which is attributed to resonant enhancement due to band-gap excitation. This behavior is largely consistent with that in the PLE. Below $E_g/2$, the TPA signal increases again, and comes closes to the middle of the YL band, indicating that the YL band has large optical nonlinearity. As shown in Fig. 1(b), PLE in the YL band is two orders of magnitude smaller than that of the band-gap excitation. However, the large transmission change observed suggests that efficient TPA can occur in the YL band.

While only carriers that allow radiative recombination in the YL band can contribute to the PLE signal, changes in transmission directly reflect absorption processes. Furthermore, because excited carriers in defect states have long lifetimes,[7] any recombination probability responsible for efficient YL should be small. Therefore, the difference between PLE and TPA is attributed to the small probability of carrier recombination in the YL band.

In another study, Miragliotta and Wickenden obtained a TPA coefficient of 1.5 cm/GW by two-photon-induced photocurrent spectroscopy, which is almost the same as the present measurement near $E_g/2$.[2] In contrast, TPA coefficients below $E_g/2$ are negligible in photocurrent measurements. In photocurrent spectroscopy, because only free carriers can contribute to the signal, it seems reasonable that TPA in the YL band is negligible.

In Fig. 3(b), the maximum occurs at $\sim$1.3 eV, where the estimated TPA coefficient is as high as 1.0 cm/GW. In the corresponding two-photon energy range, this maximum exists about 1.0 eV below the band-gap resonance, which corresponds closely to the energy of the deep levels identified 1.0 eV above the valence band. Since the deep levels are predominantly produced by structural defects,[16] the breakdown of symmetry of the lattice structure may enhance TPA cross sections in the YL band. The inhomogeneous distribution of the defects activates a number of YL bands, and the lack of symmetry generates a large number of nonlinearities, thus allowing efficient TPA even far below the band gap.

Sun *et al.* obtained TPA coefficients near the band gap that were larger than those estimated by an empirical relationship.[3] This behavior near the band gap was also observed in our TPA spectrum. Sun *et al.* explained the observed TPA enhancement in terms of exciton effects. However, based on our observed efficiency of the TPA process in the YL band, the interband TPA coefficients around $E_g/2$ are considered to be affected by the TPA in the YL band. As seen in Fig. 2(a), the contribution from TPA in the YL band begins just below the band tail, which indicates that the contribution of the TPA from the YL band will also enhance the TPA coefficients around the middle of the band gap.

In summary, PLE and transient time-resolved measurements were obtained for excitation far below the band gap of GaN. The observed change in transmission below midgap is characteristic of efficient TPA, despite the lack of a band-gap transition. The TPA spectrum clearly shows that the YL band contributes to efficient TPA.

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