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Phonon-assisted ultraviolet anti-Stokes photoluminescence from GaN film grown on Si (111) substrate

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Phonon-assisted anti-Stokes photoluminescence (ASPL) in the ultraviolet region has been observed in the GaN film grown on a Si (111) substrate. The ASPL peaks are observable only at sufficiently low temperatures. In addition, even if the photon energy is ≈318 meV below the transition energy for bound excitons, the ASPL peaks can be still observed. Based on our analysis, the donor-acceptor pairs and bound excitons have played primary roles in the generation of ASPL. Upon the absorption of photons, the ionizations of the neutral donors and neutral acceptors are assisted by longitudinal-optical phonons. © *2008 American Institute of Physics*. [DOI: 10.1063/1.3030883]

Anti-Stokes photoluminescence (ASPL) or resonant frequency upconversion is one of a few optical processes for producing the output photons with higher photon energies than the input ones. The mechanisms for ASPL can be attributed to multiphoton absorption including two-photon absorption,[1–5] Auger fountain process,[6] and the absorption of phonons.[7] Such a process has been exploited for various applications such as an implementation of a visible laser,[8] detectors, displays, and laser cooling of solids.[9,10] GaN grown on a Si substrate is particularly attractive because of its low cost, large size, and potential for the integration of GaN-based optoelectronic and high-power electronic devices with a Si-based complementary metal oxide semiconductor (CMOS) electronic devices. However, the lattice mismatch between GaN and Si substrates is sufficiently large such that the tensile strain can modify the bandgap as well as exciton binding energy.

In this letter, we present our result following our investigation of phonon-assisted ASPL in GaN/Si at low temperatures. Our sample consists of GaN film grown on a silicon (111) substrate by metal organic chemical vapor deposition. Due to the large mismatch of the lattice constants and thermal expansion coefficients between GaN and Si, GaN usually thermally cracks when being directly grown on a Si substrate. The use of AlN/GaN interlayers alleviates the cracking issue, thus allowing the growth of crack-free GaN film on a Si (111) substrate. The Si (111) substrate was precleaned to remove oxide, and the 90 nm AlN buffer layer was grown at 1100 °C. Three periods of low-temperature 50-nm-thick AlN ($T_g \sim 810$ °C) and high-temperature 135-nm-thick GaN ($T_g \sim 1060$ °C) interlayers were grown, prior to the growth of the 1-μm-thick crack-free nominally undoped GaN layer ($n \approx 4 \times 10^{16}$ cm^{-3}).

Prior to performing ASPL measurements, we first have to identify the nature of the luminescent transitions by measuring a PL spectrum with the excitation photon energy far above the bandgap, i.e., using a pump source of 208 nm (5.96 eV) and an average power of 1 mW. The PL spectrum taken at 4.2 K (see Fig. 1) is dominated by the recombination of excitons bound to neutral donors, i.e., D^0X or I_2, resulting in the transition peak at 3.448 eV. Such a transition energy is consistent with the value measured previously for GaN/Si.[11,12] However, it is lower than that for the GaN film grown on a sapphire substrate by ≈24 meV.[13] Such a discrepancy could be caused by the biaxial strain between the Si substrate and the GaN epilayers, resulting in the reduction in the bandgap and exciton transition energy for GaN. One of the three small bumps at 3.400 eV can be assigned to the donor-free hole, i.e., D^0h^+.[14] The other two at 3.358 and 3.283 eV originate from the recombination of donor-acceptor pairs (D^0A^0), respectively. All the assignments made above are further supported by the evolution of the PL spectrum as the lattice temperature is varied. Figure 2 shows the fitting result by using the Varshni formula whereby we have obtained the transition energy for I_2 at $T=0$ K to be ≈3.449 eV, $\alpha \approx 8.3 \times 10^{-4}$ eV/K, and $\beta \approx 977.4$ K. Since

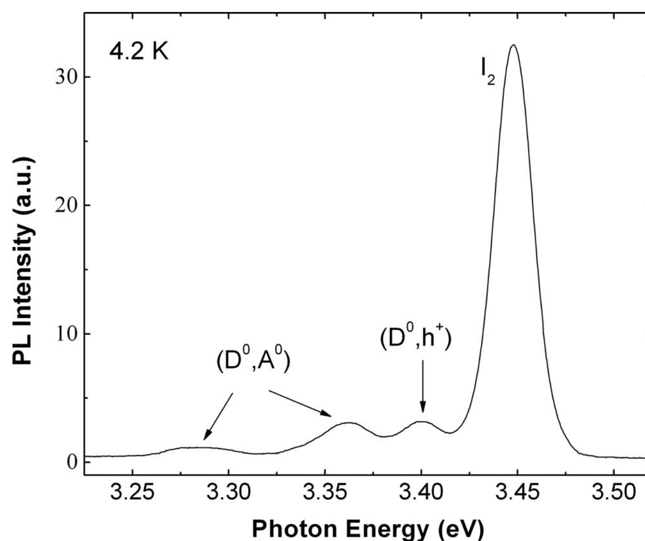

FIG. 1. PL spectrum measured at 4.2 K, photon energy of 5.96 eV, and incident power of 1 mW. Transition energies for I_2, D^0h^+, and D^0A^0 are determined to be 3.448, 3.40, 3.358, and 3.283 eV, respectively.

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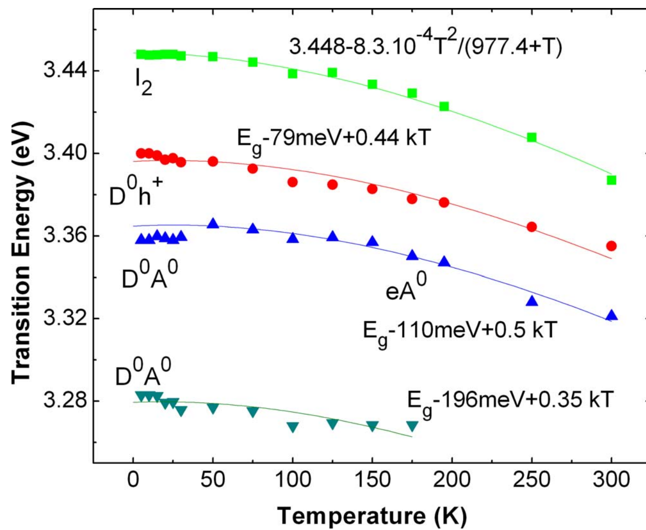

FIG. 2. (Color online) Shift of transition energy as a function of temperature for four PL peaks under the pump power of 30 mW and at a photon energy of 5.96 eV. Solid curves correspond to fitting achieved by using the Varshni relation.

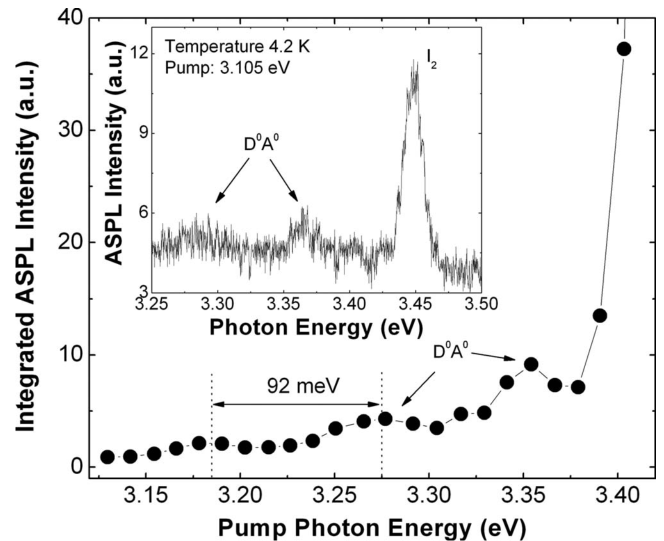

FIG. 3. PL intensity integrated over bound-exciton transition peak vs average pump power. The inset illustrates ASPL when the sample was excited at 3.105 eV with an average power of 30 mW.

the activation energy for a neutral donor in GaN is 5–7 meV,[15] the excitons bound to the neutral impurity are ionized into free excitons at temperatures higher than 100 K. At room temperature, the free-exciton transition energy is determined to be 3.387 eV according to Fig. 2. The binding energy of excitons is about 29 meV based on an Arrhenius fit to the PL intensity after being integrated over I_2 versus T^{-1}. Therefore, the bandgaps at 300 and at 0 K are 3.416 and 3.475 eV, which is 35 meV lower than the accepted value of GaN bandgap.

The temperature evolution of the transition energy for D^0h^+ can be described by[16]

$$E_{Dh} = E_g - E_D + \xi k_B T, \quad (1)$$

where E_g and E_D stand for the bandgap and binding energy for the donor, respectively, and $\xi k_B T$ originates from the temperature dependence of the transition probabilities for free holes with k_B being the Boltzmann constant. The least-squares fit to the data by using Eq. (1) yields $E_D \approx 79$ meV and $\xi \approx 0.44$, which implies that the holes participating in the transition of D^0h^+ are partially confined.[14] With increasing temperature, the donor-acceptor pair transition peak at 3.358 eV, i.e., D^0A^0, evolves to the free electron–neutral acceptor transition peak (i.e., e^-A^0) due the ionization of the donors. This is the origin for the blueshift of the transition energy near 50 K (see Fig. 2). Using Eq. (1) whereby E_D is now replaced by E_A, we have obtained $E_A \approx 110$ meV and $\xi \approx 0.50$, which indicates that the electrons participating in the recombination of e^-A^0 are partially confined. The transition peak at ≈3.283 eV has been studied in the past.[15] It is caused by the recombination of the donor-acceptor pairs, which was observed in either Mg-doped, Si-doped, or undoped GaN regardless of the substrate.

Having identified the main features of the emission spectrum, we have switched to the below-bandgap excitation by the frequency-doubled Ti:sapphire laser with a pulse width of 3 ps, a tuning range of 350–430 nm, and a repetition rate of 76 MHz. Figure 3 shows the emission spectrum at 4.2 K obtained with the pump wavelength of 399.2 nm (3.105 eV) and an average power of 30 mW. We have identified three peaks: the dominant one at 3.448 eV and two weak ones at 3.365 and 3.283 eV, respectively. According to the discussion made above, these three peaks correspond to the bound-exciton and donor-acceptor pair recombination. Also shown in Fig. 3 is the dependence of the integrated PL intensity of the bound-exciton transition peak on the pump photon energy. The steep increase of the integrated PL near 3.4 eV is the indication of the bandgap resonance for the LO-phonon-assisted absorption. Below 3.38 eV, however, the dependence in Fig. 3 just exhibits small modulations. Two tiny bumps at 3.354 and 3.277 eV correspond to the donor-acceptor pair transitions observed on the PL spectrum when the film is pumped above the bandgap (see Fig. 1). On the other hand, the transition energy for the peak at 3.185 eV is below the donor-acceptor pair transition at 3.277 eV by 92 meV, which is close to the LO phonon energy. Therefore, even when the film is pumped by the photon energies being much lower than the GaN bandgap, the bound-exciton emission is still observable, which corresponds to the ASPL.

The mechanism for ASPL observed by us can be described as follows. As a result of the ionization of the long-lived electrons bound to the donors to the conduction band upon the absorption of the photons, a large number of the nonequilibrium LO phonons are generated. These phonons have relatively long lifetime of 5 ps.[17] When the photon energy is below 3.277 eV, these phonons assist the generation of the neutral donor–neutral acceptor pairs (D^0A^0), resulting in the small peak at 3.185 eV in Fig. 3.[18] The radiative lifetime for D^0A^0 has been measured to be in the range of microseconds to milliseconds.[19] Subsequently, the photoexcitation of the electrons from the valence band to the acceptors is enhanced by the presence of these non-equilibrium LO phonons. As a result, the holes at the valence band interact with the electrons bound to the donors to form the bound excitons. When the photon energy is higher than 3.277 eV, the neutral donor–neutral acceptor pairs can be photogenerated while emitting the nonequilibrium LO phonons. These phonons are necessary to assist the generation of the bound excitons. The recombination of the bound excitons results in the I_2 peak at 3.448 eV in the inset to Fig. 3. According to

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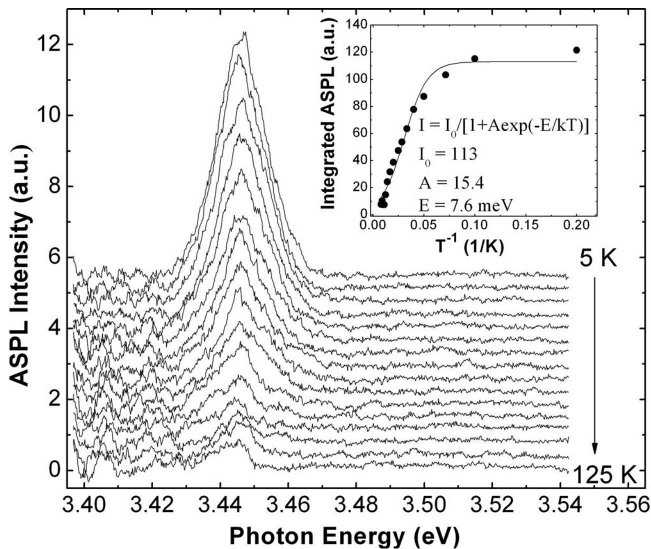

FIG. 4. Temperature dependence of PL spectra for a pump photon energy of 3.354 eV and a pump power of 30 mW. Inset: Arrhenius fit to integrated PL intensity vs reciprocal of temperature.

Fig. 4, as the temperature was increased, the ASPL peak gradually lost its strength until it disappeared. Based on the Arrhenius fit (see the inset to Fig. 4), the activation energy for I_2 is determined to be 7.6 meV. This value is in good agreement with the previous values.[15] Based on the mechanisms described above, the ASPL intensity is expected to linearly depend on the pump power. This is supported by our measurements. The rate of the increase at the photon energy of 3.356 eV is measured to be much higher. This is due to the resonance enhancement of the ASPL intensities since the photon energy is below the bound-exciton transition energy by just one LO phonon energy. Based on Ref. 20, the tensile strain being present in our sample is estimated to be $\varepsilon_{zz} \approx -0.15\%$, which reduces the GaN bandgap by 22 meV. The LO phonon energy was measured by us to be 91.7 meV,[21] which is lower than that for the GaN film grown on a sapphire substrate by just 1.6 meV.[22] Therefore, the origins of the anti-Stokes transition peaks appearing in Figs. 3 and 4 should not be affected by the amount of the strain induced in our sample.

In conclusion, we have observed an UV anti-Stokes PL from GaN film grown on a Si (111) substrate at low temperatures. The mechanism for the observed ASPL is attributed to the LO-phonon-assisted ionizations of the neutral donors and neutral acceptors, generation of neutral donor–neutral acceptor pairs, formation of the bound excitons, and subsequent recombination of these excitons. Our measurements are consistent with such a proposed mechanism. Due to the low activation energy for the bound excitons, the ASPL transition peaks are only observable at sufficiently low temperatures.

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