Photoreflectance spectra of excitonic polaritons in wurtzite GaN

Shigefusa F. CHICHIBU, Kosuke TORII[1], Takahiro DEGUCHI[2], Hisayuki NAKANISHI[2], Takayuki SOTA[1] and Shuji NAKAMURA[3,*]

Institute of Applied Physics, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8573, Tel:+81(298)53-5289, Fax:+81(298)53-5025

[1]Department of Electrical, Electronics, and Computer Engineering, Waseda University, 3-4-1 Ohkubo, Shinjuku, Tokyo 169-8555

[2]Department of Electrical Engineering, Science University of Tokyo, 2641 Yamazaki, Noda, Chiba 278-8510

[3]Department of Research and Development, Nichia Chemical Industries Ltd., 491 Oka, Kaminaka, Anan, Tokushima 774-8601

Email: chichibu@bk.tsukuba.ac.jp

Photoreflectance (PR) spectra of high-purity GaN substrate were compared with photoluminescence (PL) and reflectance spectra, which were analyzed based on a model exciton-polariton picture in which A, B, and C free excitons couple simultaneously to an electromagnetic wave. The GaN substrate with reduced threading dislocation (TD) density was prepared by lateral epitaxial overgrowth (LEO) technique during metalorganic vapor phase epitaxy (MOVPE) on sapphire with subsequent sapphire removal. The transition energies agree with the energies of bottlenecks of the excitonic polariton branches, where the lifetime of the coupled upper and lower polariton branches approach their maxima. The result means that purturbation-induced change in the dielectric function is mainly due to polaritons. Temperature dependence of the A-exciton resonance energy was well described using a model which assumes Einstein phonons.

Keywords: Exciton polariton, Photoreflectance, lateral epitaxial overgrowth (LEO).

1. Introduction

GaN and related column-III nitrides are attracting much attention as materials for optoelectronic devices like short wavelength light emitters.[1,2] The wide bandgap nature of GaN stabilizes its excitonic character even at RT, and the first observation of free exciton emissions at RT from GaN epilayers on sapphire (GaN/sapphire) has been reported[3] after Amano *et al*.[4] had observed excitonic absorption peak at RT. Recently, high quality GaN substrates have been prepared through several modifications of LEO technique,[5-7] by which nearly TD-free region was grown laterally over the patterned mask. Gil *et al*.[8] and Stepniewski *et al*.[9] have discussed exciton-polaritons in GaN/sapphire[8] and homoepitaxial layers on bulk GaN crystals grown under very high-pressure N_2 ambient.[9] Torii *et al*.[10] have also investigated low temperature reflectance and PL spectra of the GaN substrate[7] to show that the spectra were well described by a model exciton-polariton picture in which A, B, and C free excitons couple simultaneously to an electromagnetic wave. However, very little is known of the PR structure of high-quality free-standing GaN substrates. In this presentation, spectra of PR and PL in the high quality GaN substrate[7] are discussed to show that the PR method is sensitive to bottlenecks of excitonic polariton branches.

2. Experimental

The GaN substrate[7] was grown by MOVPE-LEO with subsequent sapphire removal. Detailed explanation of the preparation method and structural properties are given in Refs. 7,10 and 11. The GaN substrate exhibited biexponential-type decay with long decay constatnts of τ_1=130 ps and τ_2=860 ps at RT while a standard 1-μm-thick GaN/sapphire exhibits a short decay of τ_2=260 ps. The result means that the sample is of high purity. PR, PL and reflectance spectra were measured as a function of temperature. The spectral resolution was ±0.3 meV at a wavelength of 350 nm.

3. Results and Discussion

Figure 1 shows OR (a), PR (b), and PL (c) spectra at 10 K. Sharp reflection anomalies corresponding to the ground states of A, B, and C free excitons and the first excited states of A exciton ($A_{n=2}$) are found around 3.480, 3.485, 3.503, and 3.498 eV, respectively. The sample exhibited correspondingly sharp A, B, and $A_{n=2}$ free excitonic PL peaks and a neutral donor bound exciton peak I_2. The full width at

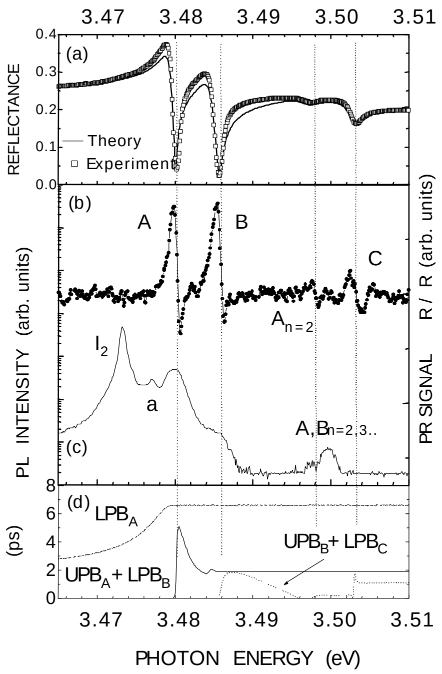

Fig.1 Optical spectra of GaN at 10 K.

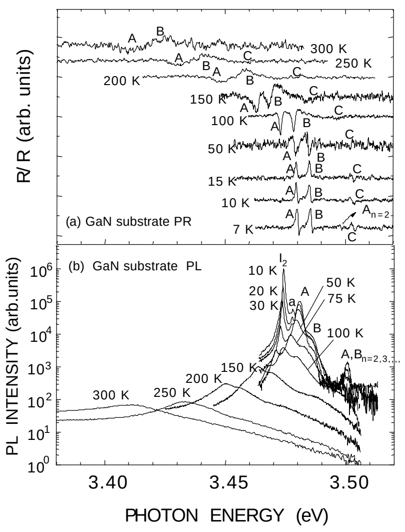

Fig.2 PR and PL spectra vs T.

half maximum (FWHM) of I_2 is as small as 0.7 meV. However, PL peaks corresponding to A and B free excitons are rather broad (2.6 meV for the A peak) and they cannot be fitted by single Lorentzian line-shape functions, suggesting the formation of excitonic polaritons. Indeed, emissions from lower and upper polariton branches of A exciton (LPB_A and UPB_A, respectively) and those of B exciton (LPB_B and UPB_B) have already been resolved using near-resonant excitation sources.[10)]

To interpret the OR spectrum, theoretical calculations have been carried out[10)] based on a model exciton-polariton picture in which A, B, and C free excitons couple simultaneously to an electromagnetic wave, where the effective mass anisotropy, the optical anisotropy, free exciton damping, and *exciton dead layer* are fully taken into account. As a result, the polariton lifetime τ has been estimated, as shown in Fig.1(d). The exciton-polariton dispersion used here has been obtained simultaneously during the calculation.[10)] The best-fit curve of $R=\left|(1-\bar{n})/(1+\bar{n})\right|^2$, where $\bar{n}$ is the effective reflactive index, is shown by a solid line in Fig.1(a). Satifactory agreement is achieved.

The PR spectrum shown in Fig.1(b) was analyzed by the low-field ER lineshape function. The transition energies are shown by dotted lines in Fig.1. Different from the PR spectra reported for unrelaxed GaN/sapphire, Γ_A of the free standing GaN is as small as 1.0 meV. From the fact that the PR transition energies agree with the energies where the lifetime of the combined exciton-polariton branches approach their maxima, i. e. bottlenecks of the excitonic polaritons in terms of the polariton dispersion, then this means that PR monitors exciton-polaritons at low temperature. It is natural to admit that PR monitors exciton-polaritons rather than excitons or free carriers since the PR signal is purely sensitive to the change in dielectric function. Thus the PL peaks corresponding to A and B transitions shown in Fig.1(c) are assigned to convolutions of upper and lower excitonic polaritons.

Fig.2 shows PR and PL spectra of the GaN substrate as a function of temperature. The PL spectra are dominated by A and B free exciton emissions above 75 K. Surprisingly, recombination of the first excited states of A excitons is recognized up to 200 K.

Transition energies obtained from the PR spectra and PL peak energies are plotted as a function of temperature in Fig.3. They show the identical temperature variation. It has been shown[12)] that in polar semiconductors that have optical phonon branches energetically next to the acoustic ones, fitting of temperature variation of a bandgap using the empirical equation proposed by Cody,[13)] which assumes Einstein phonons, is better than that using the Varshni fitting, which assumes Debye phonons. As a matter of fact, the A transition energy is fitted by the equation after Cody[13)] as $E_A(T) = 3.480\text{-}0.162[\exp(366/T)\text{-}1]$, where the A transition energy at T=0 K is 3.480 eV and Einstein characteristic temperature Θ_E is 366 K. The value of Θ_E nearly agrees with that obtained from GaN/sapphire (349 K),[12)] and this value corresponds to the energy maximum of the low-energy group of bulk phonon density of states in h-GaN (260 cm^{-1} or 370 K).[14)]

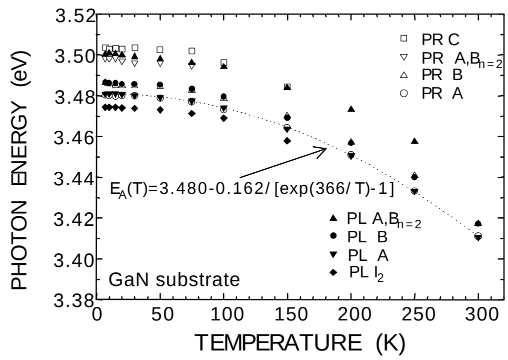

Fig.3 Transition energy vs T.

1) S. Nakamura and G. Fasol: *The Blue Laser Diode*, (Springer, Berlin, 1997). 2) I. Akasaki and H. Amano: Jpn. J. Appl. Phys., **36** (1997) 5393. 3) S. Chichibu, T. Azuhata, T. Sota and S. Nakamura: J. Appl. Phys. **79** (1996) 2784. 4) H. Amano, N. Watanabe, N. Kiode and I. Akasaki: Jpn. J. Appl. Phys., **32** (1993), L1000. 5) A. Usui, H. Sunakawa, A. Sakai and A. Yamaguchi, Jpn. J. Appl. Phys., **36** (1997) L899. 6) T. Zheleva, O-H. Nam, M. Bremser and R. Davis: Appl. Phys. Lett. **71** (1997) 2472. 7) S. Nakamura, M. Senoh, S. Nagahama, N. Iwasa, T. Yamada, T. Matsushita, H. Kiyoku, Y. Sugimoto, T. Kozaki, H. Umemoto, M. Sano and K. Chocho: Jpn. J. Appl. Phys., **37** (1998) L309. 8) B. Gil, S. Clur and O. Briot: Solid State Commun. **104** (1997) 267. 9) P. Stepniewski, K. Korona, A. Wysmolek, J. Baranowski, K. Pakula, M. Potemski, I. Gregory and S. Porowski: Phys. Rev. B **56** (1997) 15151. 10) K. Torii, T. Deguchi, T. Sota, K. Suzuki, S. Chichibu and S. Nakamura: Phys. Rev. B **60** (1999) 4723. 11) T. Deguchi, D. Ichiryu, K. Toshikawa, K. Sekiguchi, T. Sota, R. Matsuo, T. Azuhata, M. Yamaguchi, T. Yagi, S. Chichibu and S. Nakamura: J. Appl. Phys. **86** (1999) 1860. 12) S. Chichibu, T. Mizutani, T. Shioda, H. Nakanishi, T. Deguchi, T. Azuhata, T. Sota and S. Nakamura: Appl. Phys. Lett. **70** (1997) 3440. 13) G. D. Cody: Hydrogenated Amorphous Silicon, ed. J. I. Pankove, Semiconductors and Semimetals Vol.21, Pt.b (Academic, New York, 1984), Chap.2, pp.11-79. 14) T. Azuhata, T. Matsunaga, K. Shimada, K. Yoshida, T. Sota, K. Suzuki, and S. Nakamura, Physica B **219&220** (1996) 493.