PHYSICAL REVIEW B VOLUME 55, NUMBER 24 15 JUNE 1997-II

Optical properties of aluminum nitride

Qixin Guo, Mitsuhiro Nishio, and Hiroshi Ogawa
Department of Electronic Engineering, Faculty of Science and Engineering, Saga University, Saga 840, Japan

Akira Yoshida
Department of Electrical and Electronic Engineering, Toyohashi University of Technology, Toyohashi 441, Japan

(Received 25 March 1997)

Reflectance spectra measured with synchrotron radiation on AlN single crystals in the energy range from 6 to 120 eV are reported. The imaginary part of the dielectric constant for AlN was determined by using the Kramers-Kronig analysis, and the structures in the spectra were assigned based on the theoretical results reported. [S0163-1829(97)51524-4]

Aluminum nitride (AlN), one of the III-V compound semiconductors with a wurtzite crystalline structure, is promising not only for the use of passivation of semiconductor surfaces and insulators for high temperatures, but also for the use of optical devices at ultraviolet spectral region and surface acoustic wave devices, because it has the widest band gap in all the III-V compounds and high sound velocity. Recently, it has received more attention from the properties of its alloys with GaN and InN which permit the fabrication of $Al_{1-x-y}Ga_xInN_y$ based shortest-wavelength semiconductor laser diode.[1–3] In spite of the prospects of AlN in device applications, experimental data on the optical properties are surprisingly scarce,[4] although several theoretical studies were performed.[5–8] Yamashita *et al.*[9] measured the reflectance spectra of AlN in the photon energy range from 5.8 to 8.4 eV by using the deuterium lamp as the light source. Michailin *et al.*[10] attempted to measure the reflectance spectra of AlN up to the photon energy 40 eV. However, no experimental data relevant to the electronic band structures of AlN are available.[8] Accurate knowledge of the optical spectra over a wide range of wavelength is indispensable for understanding the electronic structure. In this paper, we present the results of reflectance measurements on AlN single crystals in the photon energy range from 6 to 120 eV. The imaginary part of the dielectric function derived through the Kramers-Kronig analysis are also reported.

AlN films were grown epitaxially on (0001) α-Al_2O_3 substrates using the chemical reaction of trimethylaluminum with ammonia.[11] Reflection high-energy electron diffraction results showed all the AlN samples to be a single crystal and to have an orientation relationship of (0001) AlN$\|$(0001) α-Al_2O_3. Reflectance spectra were measured at two different beam lines of synchrotron radiation facilities. One is BL1 of SOR-RING in the Institute for Solid State Physics, the University of Tokyo for the photon energy range of 6–20 eV, and another is BL5B of UVSOR in the Institute for Molecular Science for 15–120 eV. Reflectance data $R(E)$ were obtained by taking the ratio $I_r(E)/I_0(E)$, where the intensities for incident light $I_0(E)$ and reflected light $I_r(E)$ were measured in sequence in a given photon energy interval. The incident angle was nearly normal to the substrate, 15° away from the normal axis of the sample surface. Since AlN has a wurtzite structure, the reflectance data and the dielectric function are for the electric vector perpendicular to the c axis. The dielectric function was determined from the reflectance and the phase $\theta(E)$ which was calculated by the Kramers-Kronig analysis of the reflectance spectra. Since the spectral region of the reflectance measurements is always bounded ($6 \leqslant E \leqslant 120$ eV in our measurements), it becomes necessary to extrapolate $R(E)$ to infinite energies in order to estimate the phase $\theta(E)$. Below the lowest-energy limit of the measurements, the reflectance was assumed to be constant. In the higher-energy region, we used the usual typical function $R(E)=CE^{-4}$,[12] where the constant C was determined so as to connect smoothly with the measured spectrum.

Figure 1 shows a typical reflectance spectrum of AlN single crystals in the photon energy range from 6 to 120 eV. The transitions revealed at 7.9, 9.0, 13.0, and 14.8 eV are interpreted in a conventional way as the interband transitions from the valence to conduction bands. Fairly good agreement was found in the overlapped photon energy region (6.0–8.4 eV) between our reflectance spectra and those reported by Yamashita *et al.*[9] We note that our reflectance spectra differ from those reported by Michailin *et al.*[10] in the curve shape and peak positions except for the peak observed at 7.9 eV. In the higher-energy range in Fig. 1, the reflectance spectrum has a peak at 78.6 eV which is ascribed to the inner core transitions from the Al $2p$ level.

Figure 2 shows the spectral dependence of the imaginary part of the dielectric constant ε_2 for AlN. The shape of the

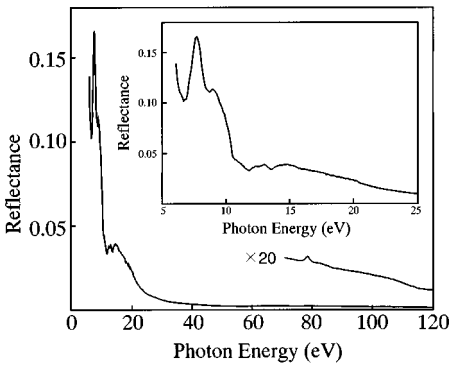

FIG. 1. Spectral dependence of the reflectance for AlN.

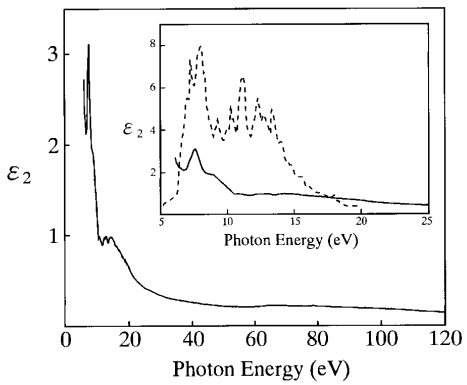

FIG. 2. Spectral dependence of the imaginary parts of the dielectric function ε_2 for AlN. The solid line indicates our experimental data, and the broken line represents the calculated result by Christensen and Gorczyca (Ref. 8).

imaginary part of the dielectric constant is similar to that of the reflectance spectrum in Fig. 1, but the actual positions of the peaks are slightly shifted in energy. There have been several calculations of band structure for AlN. Hejda[5] calculated the electronic structure of AlN by means of the orthogonalized plane-wave method. Kobayashi *et al.*[6] performed a band-structure calculation by using the semiempirical tight-binding method. Ching and Harmon[7] studied the electronic structure by means of the first-principles linear-combination-of-atomic-orbitals method. Unfortunately, our experimental data cannot be directly compared with those calculated results, because only the energy (E) vs wave-vector diagrams were reported in those papers. Recently, Christensen and Gorczyca[8] investigated the optical and structural properties of AlN by means of the linear muffin-tin-orbital method in its scalar-relativistic form in conjunction with the local-density approximation to the density-function theory. They identified the regions in k space of dominant interband contributions to the elements of structure in the dielectric function of AlN. Their calculated imaginary part of the dielectric function is presented in the inset in Fig. 2. There is a reasonable agreement between our experimental spectra and the calculated results, although the intensity of the theoretical peaks is much higher than of those in our experimental spectra because the theoretical spectra was derived in the single-particle scheme.[13] From the calculated ε_2 spectra, we assigned the peak positions for each transition, and listed them in Table I. The main peak observed at 7.6 eV in our experimental ε_2 spectra comes from transitions at the M point $(M{:}6\rightarrow 10)$. The small structure located at 8.9 eV is connected with $(6\rightarrow 10)$ transitions close to $\frac{1}{4}\Sigma R$, and the peak revealed at 13.0 eV is assigned to transitions from band 7,8 to 15,16 at the Δ point of Brillouin zone.

TABLE I. Identification of structure in $\varepsilon_2(E)$ for AlN.

Energy of optical structure (eV)		
Our results	Calculations[a]	Transition
7.6	8.08	$M{:}6\rightarrow 10$
8.9	9.35	$\frac{1}{4}\Sigma R{:}6\rightarrow 10$
13.0	13.43	Δ:7,8$\rightarrow$15,16
14.6		

[a]Reference 8.

In conclusion, we have presented optical reflectance spectra of AlN measured with synchrotron radiation in wide photon energy range from 6 to 120 eV. The imaginary part of the dielectric function for AlN was determined by using the Kramers-Kronig analysis, and each of the transitions observed has been assigned based on the linear muffin-tin-orbital calculations. The present work, complemented with theoretical studies, yields a realistic picture of the electronic band structure for AlN.

The authors would like to thank Professor K. Tsubouchi of the Research Institute of Electrical Communication, Tohoku University, for supplying the samples of AlN epitaxial films. Thanks also go to Dr. M. Fujisawa of SOR-RING facility in the Institute for Solid State Physics, the University of Tokyo and Dr. S. Kimura of UVSOR facility in the Institute for Molecular Science for their useful advice in measurements of the reflectance spectra.

[1] S. Nakamura, M. Senoh, S. Nagahama, N. Iwasa, T. Yamada, T. Matsushita, H. Kiyoku, and Y. Sugimoto, Jpn. J. Appl. Phys. **35**, L74 (1996).

[2] I. Akasaki, S. Sota, H. Sakai, T. Tanaka, M. Koike, and H. Amano, Electron. Lett. **32**, 1105 (1996).

[3] K. Itaya, M. Onomura, J. Nishio, L. Sugiura, S. Saito, M. Suzuki, J. Rennie, S. Nunoue, M. Yamamoto, H. Fujimoto, Y. Kokubun, Y. Ohaba, G. Hatakoshi, and M. Ishikawa, Jpn. J. Appl. Phys. **35**, L1315 (1996).

[4] S. Strite and H. Morkoc, J. Vac. Sci. Technol. B **10**, 1237 (1992).

[5] B. Hejda, Phys. Status Solidi **32**, 407 (1969).

[6] A. Kobayashi, O. F. Sankey, S. M. Volz, and J. D. Dow, Phys. Rev. B **28**, 935 (1983).

[7] W. Y. Ching and B. N. Harmon, Phys. Rev. B **34**, 34 (1986).

[8] N. E. Christensen and I. Gorczyca, Phys. Rev. B **50**, 4397 (1994).

[9] H. Yamashita, K. Fukui, S. Misawa, and S. Yoshida, J. Appl. Phys. **50**, 896 (1979).

[10] V. V. Michailin, V. E. Oranovskii, S. Pacesova, J. Pastrnak, and A. S. Salamatov, Phys. Status Solidi B **55**, K51 (1973).

[11] M. Morita, N. Uesugi, S. Isogai, K. Tsubouchi, and N. Mikoshiba, Jpn. J. Appl. Phys. **20**, 17 (1981).

[12] M. Cardona, Phys. Rev. **140**, A651 (1965).

[13] C. Wang and B. M. Klein, Phys. Rev. B **24**, 3417 (1981).