Band gap of corundumlike α-Ga_2O_3 determined by absorption and ellipsometry

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(Received 24 April 2017; published 18 July 2017)

The electronic structure near the band gap of the corundumlike α phase of Ga_2O_3 has been investigated by means of optical absorption and spectroscopic ellipsometry measurements in the ultraviolet (UV) range (400–190 nm). The absorption coefficient in the UV region and the imaginary part of the dielectric function exhibit two prominent absorption thresholds with wide but well-defined structures at 5.6 and 6.3 eV which have been ascribed to allowed direct transitions from crystal-field split valence bands to the conduction band. Excitonic effects with large Gaussian broadening are taken into account through the Elliott-Toyozawa model, which yields an exciton binding energy of 110 meV and direct band gaps of 5.61 and 6.44 eV. The large broadening of the absorption onset is related to the slightly indirect character of the material.

DOI: 10.1103/PhysRevMaterials.1.024604

I. INTRODUCTION

Gallium oxide (Ga_2O_3) is a wide-band-gap semiconductor which has attracted much attention over the past few years as a promising candidate for applications in a wide range of fields because of its unique physical properties. Ga_2O_3 exhibits five different polymorphs: α, β, γ, δ, and ε, of which the monoclinic β-Ga_2O_3 is the thermodynamically stable phase at ambient conditions [1]. β-Ga_2O_3 has been extensively studied as an ultraviolet (UV) transparent conducting oxide (TCO) [2,3] and a deep UV solar-blind detector [4,5]. Since the value of its refractive index is close to the square root of that of most III-V semiconductors, it is ideally suited as efficient single-layer antireflection coating for III-V semiconductors [6]. In recent years, Ga_2O_3 has been also intensively investigated for its potential applications in power electronic devices because of its large band gap ($\sim$5 eV), large breakdown field (estimated at 8 MV cm^{-1}), high static dielectric constant ($\sim$10) [6–9], and a conductivity that can be varied over a wide range [10]. Very recently, β-Ga_2O_3 Schottky barrier diodes with a breakdown voltage over 1 kV [11] and showing good performance at temperatures ranging from room temperature to 150 °C [12] have been demonstrated, which opens the possibility of practical applications of Ga_2O_3 in the next-generation power devices.

The temperature dependence of the exciton resonance has been studied by means of reflectivity [13] and a theoretical and experimental study of the complete dielectric function of β-Ga_2O_3 [14] has been reported. Very recently, it has been shown that a quasiparticle self-consistent GW calculation of β-Ga_2O_3 band structure that takes into account the lattice polarization correction gives a band gap in good agreement with experiment [15].

In addition, Ga_2O_3 may also provide low-cost substrates for GaN-based devices which combine the transparency of sapphire (α-Al_2O_3) and the conductivity of SiC. Blue light emission was observed in an InGaN multi-quantum-well structure grown by metal-organic vapor phase epitaxy (MOCVD) on β-Ga_2O_3 [16]. Homoepitaxial growth by molecular beam epitaxy of high-quality hexagonal GaN on α-Ga_2O_3 was achieved after nitridation of the α-Ga_2O_3 surface [17].

The corundumlike polymorph (α-Ga_2O_3) is a metastable phase at ambient conditions and therefore experimental studies of its basic properties are rather scarce. Nevertheless, the α-Ga_2O_3 polymorph offers new interesting possibilities of alloying with other corundum structure oxides such as Al_2O_3 and In_2O_3 for band-gap engineering [18] and also of integration of TCO technology with functional oxides such as V_2O_3, Cr_2O_3, and Fe_2O_3 to exploit multiferroic and magnetoelectric effects [19]. The growth at low temperatures (430 °C−470 °C) of high-quality single-crystalline α-Ga_2O_3 films on c-sapphire by mist chemical vapor deposition (CVD) was demonstrated by Shinohara and Fujita [20]. The fact that both the epilayer and the substrate have the same crystalline structure favors the growth of layers with excellent crystalline quality. Mist CVD is a cost-effective technique that can provide large-area, high-quality α-Ga_2O_3 layers which are susceptible to be used as buffer layers for the growth of epitaxial hexagonal GaN layers since they have the same rhombohedric crystalline structure as sapphire and an excellent lattice match of GaN to the nitridated α-Ga_2O_3 surface is expected. The availability of high-quality, mist-CVD-grown α-Ga_2O_3 films has triggered a renewed interest in the material and its fundamental optical properties. A good knowledge of the optical properties of α-Ga_2O_3 is necessary to realize the full potential of this promising material. Recently, a lattice dynamics study was carried out by means of polarized Raman scattering on a mist-CVD grown α-Ga_2O_3 single crystal [21]. A first determination of the α-Ga_2O_3 fundamental band gap (E_g) was carried out by Sinha *et al.* [22], who reported a value of $E_g = 4.98$ eV estimated from an α^2-versus-$h\nu$ plot of the absorption spectrum. Later, a value of 5.3 eV was obtained from the transmittance spectrum [20]. However, both estimates are based on the low-energy tail of the absorption

coefficient, which is well known to yield underestimated E_g values. The electronic structure of α-Ga_2O_3 was theoretically investigated by means of density functional theory in the linear combination of atomic orbitals approximation (LCAO-DFT) [23]. According to these calculations, the valence band at Γ is slightly lower in energy than at the L or F points of the Brillouin zone, which leads to an indirect L-Γ or F-Γ gap. Later calculations based on the self-consistent screened exchange local density approximation (LDA) method yielded a value of $E_g = 5.40$ eV [24]. Recently, single-quasiparticle band structure calculations based on Hedin's GW approach have been performed, and a value of 5.63 eV has been obtained for the direct gap at the Γ point [25].

In this work, we present an investigation of the absorption edge of α-Ga_2O_3 by means of transmission and ellipsometry measurements in the UV range (400–190 nm). The analysis of the absorption structures with the excitonic Elliott-Toyozawa theory [26,27] allows a reliable determination of the band gap and electronic structure near the band edge. We have found a value of $E_g = 5.61$ eV at room temperature (RT) for the corundumlike α-Ga_2O_3 films, in very good agreement with the recently calculated band gap for this compound [25]. The temperature dependence of E_g has been studied in the 6–300 K range and is well described by a simple electron-phonon interaction model based on the Bose-Einstein statistical factors for phonon emission and absorption [28]. We present also ellipsometry measurements in the 4.8–6.5 eV energy range at RT. From measurements of the imaginary part ε_2 of the dielectric function, the absorption spectrum in this energy range has been obtained and shows good agreement with the transmittance measurements.

II. EXPERIMENT

(0001) corundumlike α-Ga_2O_3 thin films of thickness ranging from 80 to 500 nm were grown by ultrasonic mist CVD on a c-face sapphire substrate. A liquid solution of $Ga(C_5H_7O_2)_3$ in H_2O (0.05 mol/L) was used as Ga source. Mist particles were formed by a 2.4-MHz ultrasonic transducer and carried by a flow of N_2 gas to the heated reaction chamber, where the (0001) sapphire substrate was placed on a sample holder kept at 200 °C. The samples were characterized by x-ray diffraction (XRD). The layers exhibited a high-quality pure corundum phase as indicated by a distinct (0006) α-Ga_2O_3 XRD reflection. The XRD rocking curves for most of the samples display full widths at half maximum below 100 arcsec.

Transmission measurements were performed in a purely reflective homemade optical set-up. A deuterium lamp was used as a source in the UV range. Aluminum mirrors were used for focusing the light beam on the sample at normal incidence, collecting the transmitted beam and focusing it on the end of an optical fiber that guides it to the entrance slit of the spectrometer. A UV multichannel spectrometer was used to analyze the reference and transmitted spectra in the 190–600 nm range. Transmission experiments at low temperature were taken, with the same optical setup, using a closed-cycle He cryogenerator with fused silica windows. Absorption spectra were obtained from the transmission spectra by assuming zero absorption in the spectral range exhibiting constant contrast interference fringes. A commercial rotating analyzer ellipsometer with autoretarder was used for the spectroscopic ellipsometry studies which yielded Ψ and Δ with high accuracy for angles of incidence between 56° and 74° in steps of 7°. Real and imaginary parts of the dielectric function were obtained via point-by-point fitting experimental data including all five angles of incidence using a multilayer model taking into account the (0001)-oriented sapphire substrate, the Ga_2O_2 layer, and a surface roughness layer approximated as a Bruggeman effective medium. The thin-film thickness has been determined from the optical path (as obtained from the reflection interference fringe pattern) and the refractive index spectrum (as obtained from ellipsometry).

III. RESULTS AND DISCUSSION

Figures 1(a), 2(a), and 2(b) show the absorption edge of α-Ga_2O_3 as obtained from the transmittance spectra of two

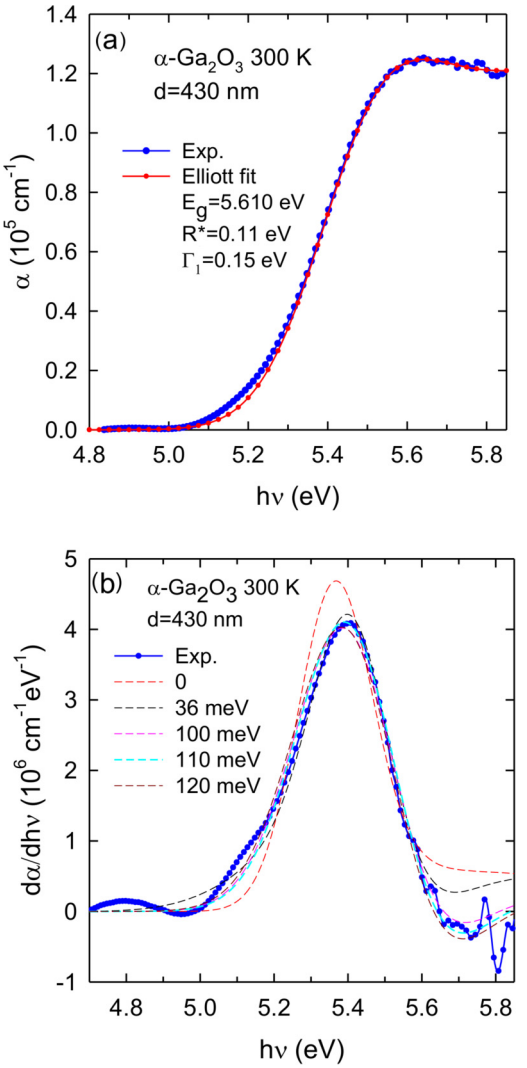

FIG. 1. (a) Experimental absorption edge of a 430-nm-thick sample of α-Ga_2O_3 at 300 K as obtained from its transmittance spectra (blue curve) and Elliott-Toyozawa fit (red curve) using the parameters shown in Table I. (b) Derivative of the experimental absorption edge with respect to the photon energy (blue solid curve) compared to the derivative of several fits using different values of the exciton ionization energy R^*.

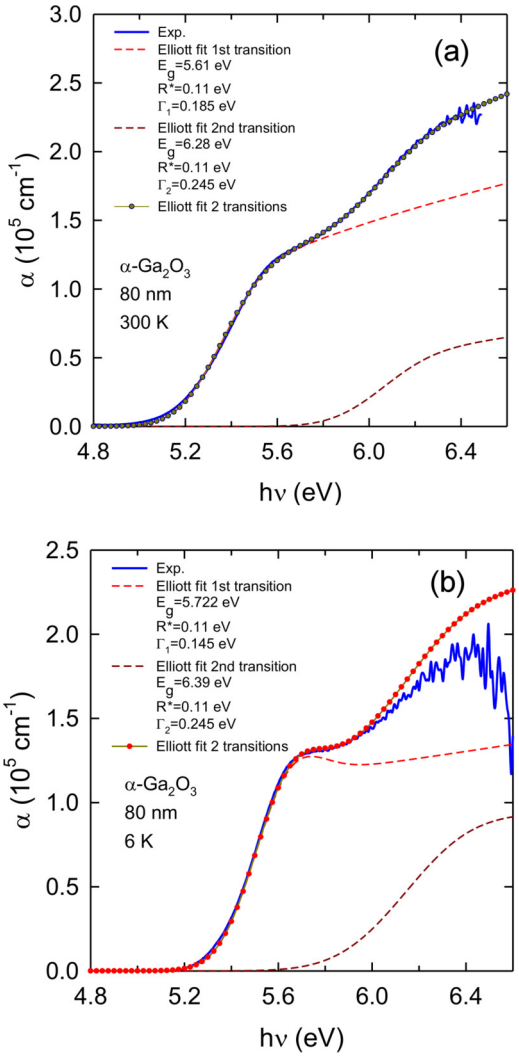

FIG. 2. (a) Experimental absorption edge of an 80-nm-thick sample of α-Ga_2O_3 at 300 K as obtained from its transmittance spectra (blue curve) and Elliott-Toyozawa fit using the parameters shown in Table I. (b) Same at 6 K. The absorption/reflection of the cryostat windows reduces the photon flux and consequently the absorption spectra beyond 6.2 eV becomes noisier.

samples with different thickness at RT and 6 K. At RT, reliable values of the absorption coefficient for the thinnest sample could be obtained up to 6.5 eV, whereas for thicker samples reliable values could be obtained only up to about 6 eV due to the lower transmittance of the layer. In the low-temperature measurements, the absorption/reflection of the cryostat windows decreases the transmitted photon flux and, consequently, further reduces the range of the absorption spectra in the UV.

The absorption edge of α-Ga_2O_3 exhibits two steplike onsets that become slightly steeper at low temperature. In the thick sample, a maximum at 5.6 eV is clearly observed both in the absorption and in the ellipsometry spectra at room temperature (see Fig. 3). These features suggest that the absorption edge of α-Ga_2O_3 arises from two allowed direct transitions in which excitonic effects are observed even at RT. This can be confirmed by fitting the spectral

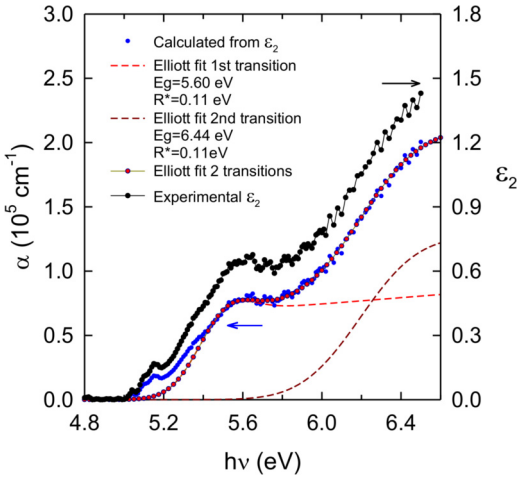

FIG. 3. Imaginary part (ε_2) of the dielectric function of α-Ga_2O_3 obtained from ellipsometry at RT on a 473-nm-thick sample (black curve). Absorption coefficient of α-Ga_2O_3 obtained from ε_2 (blue curve), compared to the Elliott-Toyozawa model fit (red and brown dashed lines) using the parameters shown in Table I [sample 4(e)].

shape obtained by means of the excitonic Elliott-Toyozawa model [26,27]. An analysis of the numerical derivative of the absorption edge shows that the transition broadening is Gaussian, indicating strong electron-phonon coupling [27]. In this case, the Elliott-Toyozawa model leads to the following expression:

$$\alpha(E) = \frac{C_0 R^{*\frac{1}{2}}}{E\sqrt{2\pi}} \left\{ \sum_{m=1}^{\infty} \frac{2R^*}{m^3} \frac{1}{\Gamma_m} \exp\left[-\frac{(E-E_m)^2}{2\Gamma_m^2} \right] + \frac{1}{\Gamma_c} \int_{E_g}^{\infty} \frac{\exp\left[-\frac{(E'-E)^2}{2\Gamma_c^2} \right]}{1 - \exp\left[-2\pi \sqrt{\frac{R^*}{E'-E_g}} \right]} dE' \right\}, \quad (1)$$

where $C_0 = [(2\mu)^{3/2} e^2 \mu_0^{1/2} |P_{CV}|^2]/(nh^2 m_0^2 \varepsilon_0^{1/2})$, $E_m = E_g - R^*/m^2$, and $\Gamma_m = \Gamma_c - (\Gamma_c - \Gamma_1)/m^2$. Here, E_g is the direct band gap, R^* is the Rydberg exciton effective energy, E_m and Γ_m are, respectively, the energy and the Gaussian width of the discrete exciton absorption peaks, and Γ_c is the Gaussian width of the exciton continuum. Figures 1(a), 2(a), and 2(b) show the fit of Eq. (1) to the experimental spectra. The values of the fitting parameters are given in Table I. It must be noted that for the thick samples a single Elliott-Toyozawa contribution is used in the fit because transmission measurements do not provide reliable values of the absorption coefficient for photon energies higher than 6 eV. For the thinnest sample (sample 2), a second contribution is added to account for the second transition. From the values of the fitting parameters we deduce a value for the band gap of α-Ga_2O_3 of 5.61 eV at RT. This result is in excellent agreement with quasiparticle band structure calculations recently reported [25].

The estimation of R^* from the RT absorption spectra is hampered by the absence of an intense exciton peak. To enhance the accuracy of the R^* estimate, we have considered the derivative of the experimental absorption edge around E_g.

TABLE I. Band-gap energy and broadening parameter for the two lowest-energy allowed transitions of α-Ga_2O_3 at 300 and 6 K as obtained by fitting the Elliott-Toyozawa model to the experimental absorption edge of several samples. A fixed value of $R^* = 0.11$ eV, determined from the absorption derivative fit shown in Fig. 1(b), was used for all samples. For sample 4, two sets of parameters are given, as the absorption edge was measured by both optical transmission [4(t)] and spectroscopic ellipsometry [4(e)].

Sample	d (nm)	T (K)	E_{g1} (eV)	Γ_1 (eV)	E_{g2} (eV)	Γ_2 (eV)
1	430	300	5.61	0.150		
2	80	300	5.60	0.180	6.28	0.245
2	80	6	5.722	0.150	6.39	0.245
3	232	300	5.62	0.162		
3	232	6	5.718	0.132		
4(t)	473	300	5.605	0.162		
4(e)	473	300	5.60	0.140	6.44	0.250

Figure 1(b) shows theoretical fits to the derivative of the absorption edge using different R^* values. The theoretical curves are obtained by numerical differentiation of Eq. (1). These indicate that the shape of the absorption derivative is very sensitive to the value of R^* employed, and therefore they allow us to reliably estimate a value of $R^* = 110$ meV for α-Ga_2O_3. The analysis indicates that the error in the estimated value of R^* is below 10%.

In most wide-gap polar materials the exciton binding energy is higher than the Rydberg exciton effective energy derived from the hydrogenic model, which is given by

$$R_0^* = \mathrm{Ry}\frac{m_x^*/m_0}{\varepsilon_s^2/\varepsilon_0^2}, \tag{2}$$

where Ry ≈ 13.6 eV is the Rydberg energy, m_x^* is the exciton reduced effective mass, and ε_s is the static dielectric constant. To our knowledge, there are no ε_s values available in the literature for α-Ga_2O_3. Therefore, we have assumed for this compound the same static dielectric constant ($\varepsilon_s = 10$) that was reported for β-Ga_2O_3 [6–9]. For the exciton reduced effective mass, given the flatness of the top of the valence band in α-Ga_2O_3, we consider the electron effective mass $m_e^* = 0.276\, m_0$ reported in Ref. [23]. Then, we obtain a value of $R_0^* \sim 36$ meV for α-Ga_2O_3. It is obvious from Fig. 1(b) that such low value of R_0^* is not consistent with the shape of the experimental absorption edge.

The large discrepancy between this estimate and the value of 110 meV obtained from our transmission measurements can be explained by the strong electron-phonon coupling that is expected in polar materials. This is determined by the electron-polar phonon coupling constant (Fröhlich coupling constant) [29] for electrons (α_e) and holes (α_h). We have performed *ab initio* calculations of the polar modes of α-Ga_2O_3 which yield an energy of 84.6 meV for the E_u(LO) mode. Considering this longitudinal optical (LO) phonon energy and $\varepsilon_s = 10$, we obtain an estimate of the electron-phonon interaction $\alpha_e = 1.054$, in the low-to-intermediate range of electron-phonon-coupling strength ($\alpha_e < 6$) where the predicted binding energy of the exciton 1s state is

$$R^* = R_0^* + \alpha_e E_{\mathrm{LO}}. \tag{3}$$

Then, from Eq. (3) we find $R^* \approx 126$ meV, which is only slightly higher than the value obtained from the fits to the absorption spectra.

The large broadening of the absorption onset is related to the specific characteristics of the valence band structure of α-Ga_2O_3 [23–25]. The top of the valence band is located about 240 meV higher than the valence band energy at the Γ point [25]. Therefore, the absorption peaks of the discrete exciton spectrum (as well as the exciton continuum absorption) are resonant with the continuum of indirect transitions from the VBM to the conduction band minimum (CBM) at the Γ point. Excitons generated in the direct transition at the Γ point have a very short lifetime because holes are quickly thermalized from the Γ point to the VBM. Even at low temperature, when phonon absorption scattering processes have a low probability, holes are quickly thermalized by phonon emission, and the exciton width remains large.

The exciton absorption dominates the spectral range from 5 to 5.6 eV and overlaps the range of indirect transitions that are expected to be several orders of magnitude weaker, which prevents the determination of the indirect band gap by optical absorption. Figure 3 shows the imaginary part (ε_2) of the dielectric function measured by ellipsometry (black curve) and the absorption spectrum of α-Ga_2O_3, as calculated from ε_2 using the equation $\alpha = 2\pi\varepsilon_2/\lambda n$ (blue curve). The refractive index, as determined from ellipsometry measurements, changes by less than 2% over the energy range studied. An average value $n = 2.35$ has been used in the calculation. The shape of the absorption curve is basically the same as in Fig. 2(a), but its amplitude turns out to be 30% lower, which may be related to surface effects and internal voids which are partly taken into account in the ellipsometry data analysis. Figure 3 also shows an Elliott-Toyozawa fit to the absorption edge calculated from ε_2. The fit parameters are shown in Table I [sample 4(e)]. It must be stressed that the small deviation of the model from the extracted absorption coefficient for very small values of ε_2 is most likely due to uncertainties in the layer model or possible contributions from indirect interband transitions which are not accounted for in the Elliot-Toyozawa model. Besides, the ellipsometry technique can yield relatively large errors for very small values of ε_2. On the other hand, the second band-gap energy obtained from the ellipsometry measurements (6.44 eV) is in principle more reliable than the one obtained from transmission measurements, as the transmitted intensity in this spectral range is very low. It should be remarked that this value is in excellent agreement with the value 6.43 eV reported as a strong interband transition in a recent quasiparticle band structure calculation [25].

In Fig. 4, the temperature dependence of E_g and of the exciton broadening are displayed between 6 and 300 K for two samples with different thickness. The temperature dependence of E_g was determined by fitting Eq. (1) to the absorption spectra at each temperature. In these calculations, a constant value of R^* was used. In actual fact, as temperature decreases from RT to 6 K, the electron effective mass, assumed to be proportional to E_g, increases by about 1.5% whereas the static dielectric constant decreases by about 2% (as determined from the shift of the transmission spectrum interference fringes). Then, the hydrogenic R_0^* contribution to the exciton binding

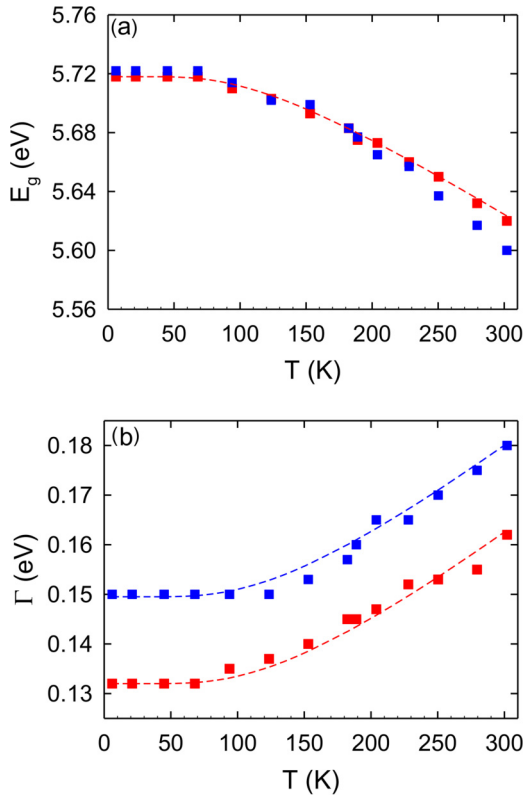

FIG. 4. Temperature dependence of the band gap (a) and transition width (b) as obtained from the Elliott-Toyozawa model fits to the experimental absorption coefficients of two samples at different temperatures. Squares correspond to the experimental data and dashed lines correspond to the fits of the model described by Eqs. (4) and (5). Blue and red symbols correspond to data from samples 2 and 3 in Table I, respectively.

energy in Eq. (2) is expected to increase by about 5%–6%. The temperature dependence of the LO phonon energy is expected to be much lower. Therefore, the change in the value of R^* in this temperature range is well below the typical error bars of the fitting procedure. The temperature dependence of E_g can be modeled by a simple electron-phonon interaction model that takes into account Bose-Einstein statistics for phonon absorption and emission rates [28]. This model is given by the equations

$$E_g(T) = E_g(0) - An_\omega \tag{4}$$

and

$$\Gamma(T) = \Gamma_0 + Bn_\omega + \Gamma_D, \tag{5}$$

where A and B are fitting parameters, $n_\omega = [\exp(\hbar\omega/k_BT) - 1]^{-1}$ is the phonon occupation factor, k_B is the Boltzmann constant, Γ_0 is the exciton broadening at very low temperature, and Γ_D is the defect contribution, which is assumed to be independent of temperature. A good fit to the experimental data is obtained by considering a phonon energy $\hbar\omega = 30$ meV, which is close to the lowest E_g mode energy (29.8 meV) of α-Ga_2O_3 reported in Ref. [21]. As can be seen in Figs. 4(a) and 4(b), where data for samples 2 and 3 are shown, Eqs. (4) and (5) describe well the temperature dependence of E_g. For instance, in the case of sample 3, once the parameter Γ_0 has been fixed using the broadening parameter of the lowest-energy transition obtained from the fit of the Elliot-Toyozawa model at low temperature, the fit of Eqs. (4) and (5) to the temperature dependence data yields the parameter values $A = 0.21$ eV, $B = 0.09$ eV, and $\Gamma_D = 0.018$ eV. The increase of the phonon contribution to the width of the exciton absorption peak between low and room temperature as well as nonhomogeneous contributions to the broadening (defects/impurities) are on the same order as those reported for other wide-band-gap semiconductors (see, for instance, Refs. [30,31]). Then, we relate the larger exciton broadening observed in α-Ga_2O_3 ($\Gamma_0 \sim 132$ meV for sample 3) to Γ-to-VBM hole thermalization through intervalley phonon scattering, which is consistent with the high density of states around the VBM predicted by all band structure calculations.

IV. CONCLUSIONS

Transmission measurements on α-Ga_2O_3 single crystals of different thicknesses have been performed in a wide temperature range from 6 to 300 K. The absorption edge curves show two steplike onsets suggesting two allowed direct transitions with excitonic effects. This assumption has been confirmed by fitting the excitonic Elliott-Toyozawa model to the experimental data. From these fits, two band-gap energies are determined at 5.61 and 6.44 eV at RT. The value of the first band gap, which is higher than experimental values previously reported, as well as the value of the second band gap are in excellent agreement with recent quasiparticle band structure calculations for α-Ga_2O_3. An exciton ionization energy of 110 meV has been obtained by fitting the first derivative of the absorption curve around E_g.

Ellipsometry measurements have been also performed in the 4.8–6.5 eV range at RT. The absorption dispersion curve found from the ellipsometry measurements shows a very similar shape to those obtained from the transmission measurements. The Elliott-Toyozawa model provides also a good fit to the absorption curve obtained from the ellipsometry measurements, and the fitting parameters are similar to those obtained from the fits to transmittance measurements. Since the transmission intensity at the high-energy range is very low, the ellipsometry measurements have allowed us to obtain a more reliable value of 6.44 eV for the energy of the second band gap.

The temperature dependence of the α-Ga_2O_3 band gap has been determined in a wide temperature range (6–300 K). The energy and the broadening of the excitonic peak associated with the fundamental band gap display a temperature dependence that is well described by an electron-phonon interaction model based on the Bose-Einstein statistics.

ACKNOWLEDGMENTS

This work has been supported by the Spanish MINECO/FEDER under Contracts No. MAT2015-71035-R and No. MAT2016-75586-C4-1-P. We acknowledge T. Yamaguchi from the Department of Electrical Engineering and Electronics of Kogakuin University, Tokyo, Japan, for supplying us with the samples used in this study.

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