Measurement and Calculation of Optical Band Gap of Chromium Aluminum Oxide Films

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The optical band gap is a basic property of optical materials. The measured band gap depends not only on the material but also on its characteristics such as crystallinity and stoichiometry. The optical band gap of chromium aluminum oxide films was measured and calculated by three different methods. Firstly, we used the conventional experimental-graphical method, which is commonly used. However, this method is applicable only to an all-crystalline phase or an all-amorphous phase. The second one was an experimental-calculation method applicable to films composed of both crystalline and amorphous phases. We calculated the optical band gap between the highest occupied molecular orbital of O2p and the lowest unoccupied molecular orbital of Cr3d in $Cr_{1.71}Al_{0.29}O_3$ films composed of both amorphous and crystalline phases. Also, a band gap for the d–d transition was obtained. Finally, the measured value was compared with the theoretical optical band gap calculated by the discrete variational-Xα (DV-Xα) method.

KEYWORDS: chromium aluminum oxide, optical band gap, amorphous, crystalline, film, measurement, calculation

1. Introduction

Chromium aluminum oxide has been proposed as a candidate material for deep-ultraviolet (DUV) optical masks.[1] In terms of the applications, optical properties were correlated to the structural properties, and a rather simple calculation procedure enabled us to find an optimum condition for mask fabrication. However, we thought that it would be better to study the relationship between the optical and the structural properties than to describe the film property only empirically. Therefore, we attempted to calculate the optical band gap of films consisting of both crystalline and amorphous phases to gain insight into its relation with the structural properties. A similar study of the optical band gap of the mixed phases was performed previously for chromium oxide thin films.[2]

Two kinds of band gaps were investigated in this research. One is the gap E_{p-d} between the highest occupied molecular orbital of O2p and the lowest unoccupied molecular orbital of Cr3d, which is commonly accepted as the optical band gap. Another is the band gap E_{d-d} for the d–d transition, which is located just below E_{p-d} in the energy scale.[3]

First of all, we followed the approach of the conventional method to measure the optical band gap of chromium aluminum oxide films deposited by sputtering. Since perfect amorphous films are difficult to fabricate by sputtering due to the high energy associated with sputtered ions and the thermodynamic stability of the crystalline phases,[4] it is likely that both the crystalline phase and amorphous phase form in the sputtering process. However, the conventional method supposes ideal cases of either a pure amorphous or a pure crystalline phase. Thus, it seems difficult to apply this approach to the calculation of the combined mixture of the crystalline phase and amorphous phase. A large discrepancy in the calculation of optical band gaps measured for the same sample using the two different assumptions of pure amorphous and pure crystalline phases in the analysis supports our idea. Therefore, the optical band gap was calculated from an analysis suggested by Hong *et al.* (experimental-calculation method),[2] which is applicable to the combined mixture of the crystalline phase and amorphous phase in the film. The mea-

sured value was compared with the theoretical optical band gap of chromium aluminum oxide calculated with a DV-Xα simulator.[5]

2. Theoretical Background

2.1 Optical transition

Basically, there are two types of optical transition that can occur at the fundamental edge of crystalline semiconductors: direct and indirect transitions. Both involve the interaction of an electromagnetic wave with an electron in the valence band, which is raised across the fundamental gap to the conduction band. However, indirect transitions also involve simultaneous interaction with lattice vibrations. Thus the wave vector of the electron can change in the optical transition, the momentum change being taken or given up by phonons.[6]

If exciton formation (electron-hole interaction) is neglected, the forms of the absorption coefficient α as a function of photon energy $h\nu$ depend on the dependence on energy of $N(E)$ of the bands containing the initial and final states. For simple parabolic bands ($N(E) \propto E^{1/2}$) and for direct transitions,

$$\alpha(\nu)n_0 h\nu \approx (h\nu - E_g)^n, \qquad (1)$$

where n is a constant of 1/2 for allowed transitions and of 3/2 for forbidden transitions in the quantum-mechanical sense. n_0 is the refractive index which is assumed to be a constant over energy variation, $h\nu$ is the photon energy and E_g is the band gap energy of the material under investigation. This type of absorption, shown in Figs. 1(a) and 1(b), is independent of temperature apart from any variation in E_g. For indirect transitions,

$$\alpha(\nu)n_0 h\nu = \frac{(h\nu - E_g + h\nu_{ph})^n}{\exp(h\nu_{ph}/kT) - 1} + \frac{(h\nu - E_g - h\nu_{ph})^n}{1 - \exp(-h\nu_{ph}/kT)}. \qquad (2)$$

The two terms here represent contributions from transitions involving phonon absorption and emission, and different coefficients of proportionality and temperature dependences. For allowed transitions $n = 2$ and for forbidden transitions $n = 3$. In each case, multiple-phonon processes can occur, leading to additional pairs of terms.

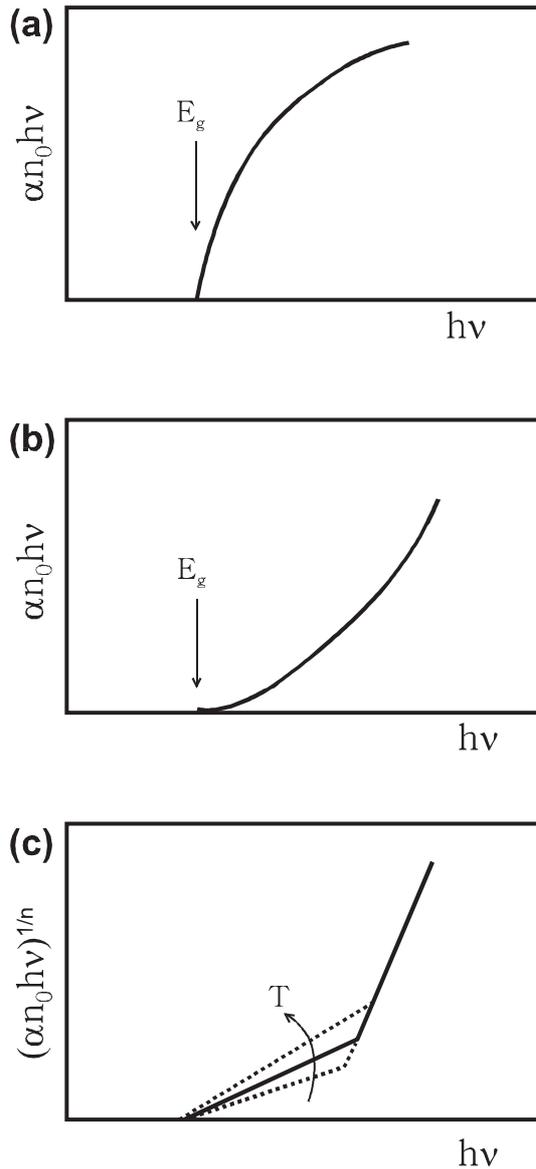Equation (1) can also be applied to amorphous materials

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Fig. 1. Schematic illustration of different kinds of absorption edges in crystalline semiconductors: (a) direct allowed, (b) direct forbidden, and (c) indirect transition.

if the value of n is 2.[7] The transition is characterized by a "nondirect" transition that is different from an "indirect" transition in the sense that it does not involve phonon absorption or emission during the transition. However, since the transition occurs nonvertically in the E–k diagram, it is also different from a "direct" transition where the k conservation rule applies. Tauc and Mott independently derived the equation for the optical band transition of amorphous materials and it was found that the equation has the same form as eq. (1) with $n = 2$.[6,7]

2.2 Experimental-calculation method of band gap measurement

The experimental-calculation method was proposed by Hong *et al.*[2] to estimate the optical band gap of a film composed of both crystalline and amorphous phases. In this method, several assumptions were taken into account. First, the optical band gaps of the amorphous phase and of its corresponding crystalline phase were assumed to have a common

optical band gap E_g. In principle, the chemical bonding characteristics of the amorphous phase differ from those of the crystalline phase. However E_g can be treated as a representative value representing the optical band gap of the mixture of the amorphous phase and crystalline phase. Secondly, the structure of the film is assumed to be multilayered, each layer being exclusively amorphous or crystalline. This assumption is supported by direct observation of the multilayered structure by a transmission electron microscope; more information is available in ref. 2. Finally, this method is applied to direct allowed transition in crystalline phases and to nondirect transition in amorphous phases near the fundamental band gap.

With these assumptions, it can be speculated that the optical density δ or $\alpha \Delta d$ is expressed as[8]

$$\delta = f_1 \delta_c + (1 - f_1)\delta_a, \tag{3}$$

where f_1 is the relative fraction of the crystalline phase and subscripts 'c' and 'a' denote crystalline and amorphous, respectively. According to the theories relevant to the optical band, the optical density for each phase should be

$$\delta_c h\nu = C_1(h\nu - E_g)^{\frac{1}{2}}, \tag{4}$$

$$\delta_a h\nu = C_2(h\nu - E_g)^2, \tag{5}$$

where h is the Planck constant, ν is frequency of the incident light, and C_1 and C_2 are constants. If we denote $h\nu$ as x and $h\nu$-E_g as M for convenience and rewrite eq. (3) in combination with eqs. (4) and (5), eq. (3) becomes

$$\delta x = f_1 C_1 M^{\frac{1}{2}} + (1 - f_1)C_2 M^2. \tag{6}$$

Differentiating eq. (6) with respect to x once and twice yields

$$x\frac{d\delta}{dx} + \delta = \frac{1}{2}f_1 C_1 M^{-\frac{1}{2}} + 2(1 - f_1)C_2 M \tag{7}$$

and

$$x\frac{d^2\delta}{dx^2} + 2\frac{d\delta}{dx} = -\frac{1}{4}f_1 C_1 M^{-\frac{3}{2}} + 2(1 - f_1)C_2. \tag{8}$$

Rearranging eqs. (6)–(8) with respect to M, we can obtain the quadratic equation

$$2(x\delta'' + 2\delta')M^2 - 3(\delta + x\delta')M + 2x\delta = 0, \tag{9}$$

where δ' and δ'' are the first and the second derivatives of δ with respect to x. Equation (9) gives two possible solutions for M with respect to x. Therefore, there also exist two possible solutions for E_g. Among the two solutions of eq. (9), the solution showing the larger value for E_g is not suitable experimentally. Therefore, the following solution of eq. (9) is chosen for E_g.

$$E_g = x - \frac{3(\delta + x\delta') + \sqrt{9(\delta + x\delta')^2 - 16x\delta(x\delta'' + 2\delta')}}{4(x\delta'' + 2\delta')}$$

$$= x - \frac{3(\alpha + x\alpha') + \sqrt{9(\alpha + x\alpha')^2 - 16x\alpha(x\alpha'' + 2\alpha')}}{4(x\alpha'' + 2\alpha')} \tag{10}$$

2.3 DV-Xα method

There are several methods to investigate the electronic band structure or energy state of semiconductors and insulators theoretically. The DV-Xα method is a successful method for studying a transition-metal compound.[9] The DV-Xα method

is a linear combination of atomic orbital-molecular orbital calculations. Adachi and Tsukada[5] outlined the basic theory and also fully provided its description. One of the advantages of the DV-Xα cluster method is that the electronic states and energy level distributions can be provided in a relatively short computing time not only for simple molecules but also for metal complexes or large clusters which may contain many transition metal atoms. Also, this method has the merit of using the Slater exchange potential and correlation energies.[10]

3. Experimental Procedures

Chromium aluminum oxide films were deposited using a DC magnetron reactive sputtering system with a disc-shaped 4 inch Cr–Al(16 at.%) target (3N).[1] Inside the chamber, we introduced a grounded mesh to improve film uniformity and quality.[11] The target was presputtered for 20 min with Ar plasma. Then the Ar and O_2 gas mixture was injected into the chamber, with flow rates of 2 sccm (1 sccm = 1 atm cm^3/min = 2.68×10^{19} mol/min at standard condition) and 1 sccm, respectively. The films were deposited under the following conditions: substrate temperature of 300°C, power of 40 W and total chamber pressure of 4.7×10^{-3} Torr. The target to substrate distance was fixed at 132.4 mm. Films were deposited on quartz and Si substrates simultaneously. Film thickness was measured from cross-sectional scanning electron microscope (SEM) images. Film composition was analyzed by a wavelength dispersive spectrometer (WDS). The phase analysis was performed by the X-ray diffraction (XRD) method. A 212-nm-thick film was deposited under the same conditions as used for the above films to observe X-ray crystalline peak and the peak was set as a reference to identify otherwise unclear peaks observed in the much thinner films described in Table I.

Before calculating the band gaps, we confirmed whether the chromium aluminum oxide films show direct or indirect transition. We examined the dependence of the transmission and absorption spectra on the temperature because the indirect transition has a large temperature dependence due to phonon involvement whereas the direct transition shows negligible temperature dependence. The transmittance spectrum was measured by UV-visible spectroscopy in the wavelength range from 222 nm to 550 nm. During the measurement, the film temperature was varied from 26°C to 72°C. Film temperature was measured using a surface thermometer.

After checking the nature of the transitions for chromium aluminum oxide films, we measured and calculated the optical band gaps.   From the measured transmittance

Table I.   Properties of chromium aluminum oxide films.

	Film 1	Film 2
Thickness (nm)	111	138
Al : Cr (in at.%)	14.9 : 85.1	14.4 : 85.6

Table II.   Crystal structure parameters of Cr_2O_3 and Al_2O_3.

	System	Space group	a (nm)	c (nm)	c/a
Al_2O_3	Rhombohedral	D_{3d}^6	0.4785	1.2990	2.72
Cr_2O_3	Rhombohedral	D_{3d}^6	0.495	1.3584	2.744

data, we obtained the optical band gap by the conventional experimental-graphical method[7] and the experimental-calculation method.[2] Also, to obtain the theoretical optical band gap, we used the DV-Xα cluster method.[5] The lattice parameters used for Cr_2O_3 and Al_2O_3 are listed in Table II.

4. Results

4.1 Microstructural characterization of films

Two films were fabricated by a DC magnetron sputtering system at 300°C to measure the optical band gap. The thickness and composition of the films are listed in Table I. The thickness difference between the two films is 27 nm. As listed in the table, they have a similar composition of about 14.6 atomic percent Al in metals (chromium plus aluminum). Figures 2(a) and 2(b) show the XRD patterns of the two films. The peak at 36.2° of 2θ corresponds to the Cr_2O_3 (110) peak.[12] As mentioned in §3, a 212-nm-thick film was deposited to obtain the standard crystalline peak for the films used in the study, and its XRD pattern is shown in Fig. 2(c).

4.2 Characterization of optical band gap transition

Before the measurement of the optical band gaps, we confirmed experimentally that chromium aluminum oxide films show direct transition. Materials showing direct transition have temperature-independent transmittance, as explained in §2. The transmittance data of the films in the wavelength range of 222 nm to 550 nm at two temperatures are shown in Fig. 3. Figure 3(a) is measured at 26°C and Fig. 3(b) is measured at 72°C. The results of (a) and (b) show nearly the same transmittance. We found that chromium aluminum oxide films have direct optical transition across the fundamental gap from the experimental finding that the transmittance spec-

Fig. 2.   XRD patterns of $Cr_{1.71}Al_{0.29}O_3$ films with thicknesses of (a) 111 nm, (b) 138 nm, and (c) 212 nm.
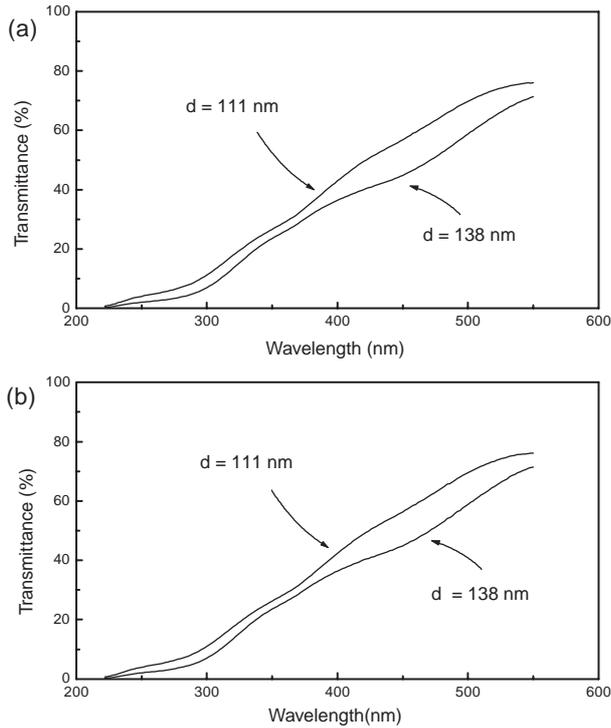
Jpn. J. Appl. Phys. Vol. 39 (2000) Pt. 1, No. 8

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Fig. 3.   Transmittance of $Cr_{1.71}Al_{0.29}O_3$ films of different thicknesses measured at (a) 26°C and (b) 72°C.

Fig. 4.   (a) $(\alpha h\nu)^2$ vs. $h\nu$ for $Cr_{1.71}Al_{0.29}O_3$ film: E_{p-d} is 5.00 eV and E_{d-d} is 3.80 eV. (b) $(\alpha h\nu)^{1/2}$ vs. $h\nu$ for $Cr_{1.71}Al_{0.29}O_3$ film: E_{p-d} is 3.90 eV and E_{d-d} is 3.18 eV.

trum is independent of temperature.

4.3   Optical band gap
4.3.1   Conventional method

Because we verified the direct transition property of chromium aluminum oxide films, we can apply eq. (1) with $n = 1/2$ (which can be applied to direct allowed transition) in the case of the crystalline film, and with $n = 2$ (which can be applied to amorphous film) in the case of the amorphous film.

Before eq. (1) is applied, the absorption coefficient can be calculated by the transmittance data of the two films, which is shown in Fig. 3(a):

$$\alpha = \left(\frac{1}{\Delta d}\right) \ln\left(\frac{T_1}{T_2}\right), \tag{11}$$

where Δd $(=d_2 - d_1)$ is the thickness difference of the two films and T_i is the transmittance of film i ($i = 1, 2$). Here, α can also be obtained by direct measurement with a spectroscopic ellipsometer.

By the graphical method based on eqs. (1) and (11), we can measure the optical band gap using the transmittance data of the two films in the wavelength range of interest and the thickness difference data. In our case, Δd is 27 nm (see §4.1). With the transmittance data [Fig. 3(a)], we measured the optical band gap. From the graphical tangent line of Fig. 4(a), we obtain 5.00 eV for E_{p-d} if we assume that the films are all crystalline. In contrast, if we assume that the films are all amorphous, we obtain 3.90 eV for E_{p-d} [Fig. 4(b)]. In the same way, we obtained E_{d-d} as shown in Fig. 4. The value of E_{d-d} is 3.80 eV if the films are assumed to be all crystalline, and 3.18 eV if the films are assumed to be all amorphous.

4.3.2   Experimental-calculation method (modified conventional method)

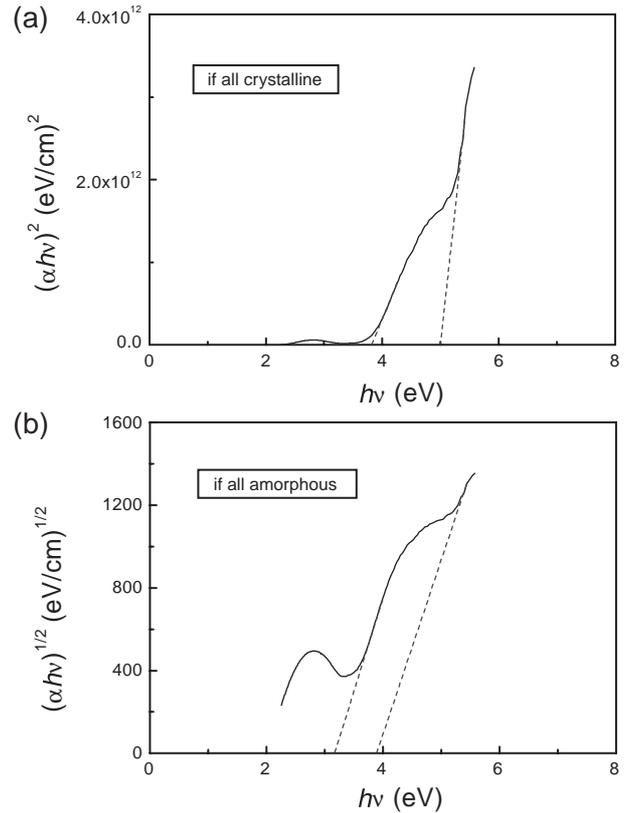Because our films are composed of both crystalline and amorphous phases, we used the experimental-calculation method[2] to measure the optical band gap. Using eq. (10), we obtained 4.65 ± 0.46 eV for E_{p-d}. In this calculation, we chose the x range (nearly no E_g variation with x) as 4.96–5.58 eV. This range corresponds to the conventional graphical tangent line range in Fig. 4. In the same way, we calculated E_{d-d} by this method. We obtained 3.73 ± 0.14 eV in the x range of 3.80–4.33 eV.

4.3.3   DV-Xα method

In this study, the DV-Xα method was employed to calculate the theoretical band gap of chromium aluminum oxide[9,13] which was compared with the measured E_{p-d} in §4.3.2. Four different Cr–Al–O cluster models containing up to 8 metal atoms were selected for the calculation. All cluster models were cut from crystalline Cr_2O_3 (or Al_2O_3) and the surfaces were terminated with oxygen atoms in order to stabilize the cluster system. The cluster models are described as follows:

Model-I: M_2–O_9;   Model-II: M_4–O_{16};

Model-III: M_6–O_{23};   Model-IV: M_8–O_{32},

where M=Cr or Al. In order to illustrate these models clearly, as examples, we plot Model-I, Model-II and Model-III in Fig. 5. The band gaps of chromium oxide and chromium aluminum oxide were calculated considering O2p to Cr3d transition. For the aluminum oxide, we considered O2p to Al3s transition. The cluster models, equivalent molecular formulas, and the calculated band gap values are listed in Table III. Figure 6 shows the variation of optical band gap E_{p-d} of chromium aluminum oxide with film composition. In §4.1,
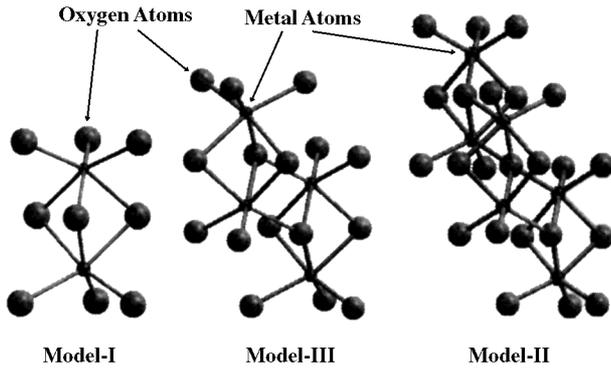
Fig. 5. Model clusters used for DV-Xα calculations; the small black spheres indicate metal atoms (Al or Cr).

Table III. Optical band gaps of chromium aluminum oxide films calculated by DV-Xα method.

Cluster model	Equivalent molecular formula	E_g (O2p → Cr3d)
Al_8O_{32}	Al_2O_3	$(9.98\,eV)^{a)}$
$Al_6Cr_2O_{32}$	$(Al_{0.75}Cr_{0.25})_2O_3$	5.21 eV
$Al_4Cr_2O_{23}$	$(Al_{0.66}Cr_{0.33})_2O_3$	4.78 eV
$Al_2Cr_4O_{23}$	$(Al_{0.33}Cr_{0.66})_2O_3$	4.43 eV
Cr_8O_{32}	Cr_2O_3	4.28 eV

a) O2p → Al3s transition.
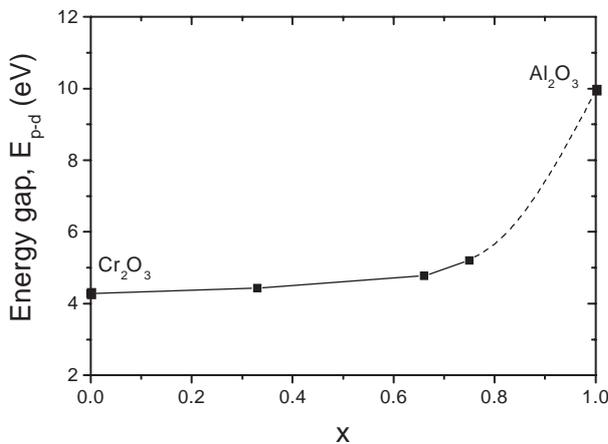

Fig. 6. Optical band gaps ($E_{p–d}$) of $Cr_{(2-2x)}Al_{2x}O_3$ calculated by the DV-Xα method.

we averaged the Al : Cr composition ratio of our films as 14.6 at.% : 85.4 at.%. The calculated optical band gap applicable to this composition yields 4.35 eV.

5. Discussion

In §4.1, we analyzed the film properties. Two films of $Cr_{1.71}Al_{0.29}O_3$ had a thickness difference of 27 nm. The crystallinity of these two films was studied by the XRD method with an incidence angle of 2°. Some studies[14–16] related to the crystallization of amorphous materials suggest that the film crystallization occurs locally so that there exists a temperature range where crystalline and amorphous phases coexist. With only XRD data, one cannot decide whether the film is composed of crystalline or amorphous phases, or both. However, in our XRD patterns, the appearance of a small broad peak, which is denoted by an arrow in Figs. 2(a) and

2(b), led us to deduce that our films are composed of both crystalline and amorphous phases. As mentioned in §4.1, a Cr_2O_3 crystalline phase was identified in the films. The amount of Cr_2O_3 was not clear, but the isolation of Cr_2O_3 may affect the composition of the remaining amorphous phase; that is, the enrichment of Al. It is better to consider this composition effect to calculate the exact band gap value. However in this study, this effect does not affect the calculation result because we assumed that the crystalline phase and amorphous phase have a common optical band gap and that it depends on the average film composition.

Before measuring optical band gaps, we confirmed experimentally that chromium aluminum oxide films show direct transition. Based on the experimental result that the transmittance spectrum is independent of temperature, chromium aluminum oxide films were found to have direct optical transition across the fundamental gap. Obtained band gaps of $Cr_{1.71}Al_{0.29}O_3$ films are summarized in Table IV. In all cases of measuring $E_{p–d}$ or $E_{d–d}$, the two values obtained with two different assumptions had a large difference. This big difference in the measured optical band gaps does not allow us to use either assumption for our films. In other words, neither assumption rigorously describes our films, thus the value obtained by either method is not reliable. This is because our films are composed of both crystalline and amorphous phases, which can be inferred from the XRD data in §4.1.

Therefore, we needed to measure the optical band gap using a modified method (experimental-calculation method) suitable for our films, as described in §4.3.2. The results of this method are summarized in Table IV with other results. As can be seen in the table, the standard deviation is much smaller in the $E_{d–d}$ calculation than in the $E_{p–d}$ calculation because of the smaller fluctuation in the transmittance data. Therefore, the value obtained by this method is more reliable when the transmittance data show less fluctuation. By comparing the two values $E_{p–d}$ and $E_{d–d}$ with the values measured by the conventional method (see §4.3.1), we found that the band gaps of our films are similar to the band gaps acquired under the assumption of an all-crystalline film. These results indicate that our films have a similar property to the crystalline films in the short range order of atoms. Also, it is inferred that chromium aluminum oxide having an Al/Cr ratio of less than 14.6/85.4 has a similar optical band gap to that of chromium oxide based on the data of Kirby *et al.*[3] who found $E_{p–d}$ and $E_{d–d}$ of chromium oxide to be 4.97 eV and 3.5 eV, respectively.

In this study, the DV-Xα method was employed to calculate the theoretical band gap of chromium aluminum oxide and to perform comparisons with the $E_{p–d}$ measured by the calcu-

Table IV. Band gaps obtained by the conventional method, experimental-calculation method, and DV-Xα method.

Method	$E_{p–d}$ (eV)	$E_{d–d}$ (eV)
Conventional method (assumption: all-crystalline)	5.00	3.80
Conventional method (assumption: all-amorphous)	3.90	3.18
Experimental-calculation method	4.65 ± 0.46	3.73 ± 0.14
x range in eV	[4.96–5.58]	[3.80–4.33]
DV-Xα method	4.35	—

lation method. With 6.9% difference, this value shows good agreement with the value obtained by the calculation method listed in Table IV.

6. Summary

Optical band gaps of $Cr_{1.71}Al_{0.29}O_3$ films were measured and calculated. By the conventional method, we obtained E_{p-d} of 5.00 eV under the assumption that all the films consisted of a crystalline phase, and 3.90 eV under the assumption that all the films consisted of an amorphous phase. Also, E_{d-d} was found to be 3.80 eV under the assumption that all the films consisted of a crystalline phase, and 3.18 eV under the assumption that all the films consisted of an amorphous phase. However, the method was not suitable to calculate the band gap by the conventional method, because our films were composed of both amorphous and crystalline phases. Therefore, we used the experimental-calculation method, which is applicable to the films composed of both the crystalline and amorphous phases. Using the experimental-calculation method, we measured the band gap of E_{p-d} as 4.65 eV, and that of E_{d-d} as 3.73 eV. These results revealed that our films have a similar property to the crystalline phase in the short range order of atoms. Finally, the E_{p-d} value was compared with the calculated value by DV-Xα, 4.35 eV.

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