Optical dispersion of Al$_x$Ga$_{1-x}$As at crystal growth temperature

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We propose a scheme to calculate the optical dispersion of III-V compounds at crystal growth temperature, based on the Afromowitz' formula and the shift of the lowest direct band gap energy. The thermal behaviors of the major parameters are extracted from their respective physical meanings. This model is applied to Al$_x$Ga$_{1-x}$As materials and the calculated results compare quite well with the experimental data. The validity of our model is also tested by comparing the static dielectric constant of GaAs with that of known empirical formula in the temperature range from 200 to 600 K. © 1994 American Institute of Physics.

I. INTRODUCTION

In situ thickness monitoring of the epitaxial growth process is necessary for growing the precise structures, such as quantum wells, superlattices, and distributed Bragg reflectors. Reflection high-energy electron diffraction is widely used as an in situ growth rate calibration method for molecular beam epitaxy, but this method can be implemented only in a high vacuum condition. Recently, with the merit of pressure independence, all-optical thickness monitoring methods such as ellipsometry and laser reflectometry are applied in chemical beam epitaxy or metalorganic chemical vapor deposition. Especially, it was shown that the distributed Bragg reflectors for a vertical-cavity surface emitting laser can be grown precisely and repeatably with the help of laser reflectometry.

Laser reflectometry is a simple method that counts the frequency of interference patterns which appear whenever the optical thickness of a thin growing layer is an integral multiple of a half wavelength of a monitoring laser. Therefore, to use the laser reflectometry as an in situ monitoring method, the knowledge about the refractive index of growing materials at growth temperatures is needed. Previous researchers calibrated these quantities by measuring the thickness of grown layers by scanning electron microscopy (SEM). However, this calibration method has some shortcomings. Firstly, both the meter-stick measurement and the ruler of SEM have several percentages of error inherently. Second, whenever the monitoring wavelength is changed, another calibration process shall be needed at the new wavelength. Therefore, the better experimental calibration method and/or a model to calculate the refractive index at crystal growth temperatures are requested. In this article we propose a high temperature version of Afromowitz' model to calculate the optical dispersion of III-V compounds at crystal growth temperature. We applied this model to AlGaAs materials and compared the calculated results with experimental data of ours and others.

II. THEORY

The real part of a refractive index can be obtained from the imaginary part of a dielectric constant through the Kramers–Kronig relation. For III-V compounds, Afromowitz proposed the imaginary part of a dielectric constant as

$$\varepsilon_2(E) = \begin{cases} \eta E^4 & E_g < E < E_f \\ 0 & \text{otherwise} \end{cases}$$  \hspace{1cm} (1)

where $E_g$ is the lowest direct band gap energy and both the $\eta$ and $E_f$ are fitting parameters. Except for the spectral region quite close to the band edge, this simple form of dielectric constant supplies quite accurate values of refractive indices over a wide spectral range. Especially, for the wavelength longer than the band gap wavelength, this model works extremely well and therefore was used by many researchers.

With this form of $\varepsilon_2(E)$, the real part of dielectric constant is written as

$$\varepsilon_1(E) = 1 + \frac{2}{\pi} \int_{E_g}^{\infty} \frac{\varepsilon_2(E')}{E'^2 - E_g^2} dE'$$ \hspace{1cm} (2)

$$= 1 + M_{-1} + M_{-3} \frac{E_f^2 - E_g^2}{E_g^2} \ln \left( \frac{E_f^2 - E_g^2}{E_g^2} \right) E_g^2, \hspace{1cm} (3)$$

where

$$M_{-1} = \frac{\eta}{2\pi} (E_f^2 - E_g^2), \hspace{1cm} M_{-3} = \frac{\eta}{4\pi} (E_f^2 - E_g^2). \hspace{1cm} (4)$$

$E_f$ and $\eta$ are obtained by comparing $M_{-1}$ and $M_{-3}$ with those of a single oscillator model which has two experimental parameters $E_0$ and $E_d$, as

$$M_{-1} = \frac{E_d}{E_0}, \hspace{1cm} M_{-3} = \frac{E_d}{E_0^3}. \hspace{1cm} (5)$$

In the single oscillator model, $E_0$ and $E_d$ have the meanings of the average interband transition energy and the oscillator strength of interband transitions, respectively. In fact, Afromowitz' model has three parameters $E_g$, $E_0$, and $E_d$ with their physical meanings.

To generate a high temperature version of the Afromowitz' model, we assumed that $\varepsilon_2(E)$ shifts with temperature at the same rate as the lowest direct band gap energy, without changing its functional form. This approximation is plausible to the first order, because the linear temperature coefficients
are all negative and nearly the same at all the interband critical-point energies of GaAs. In other words, $\varepsilon_2(E,T)$ can be written simply as
\[
\varepsilon_2(E,T) = f[E - E_g(T)].
\]
From Eq. (6), it is clear that the Kramers–Kronig integration yields the refractive indices which include the temperature as an implicit variable through $E_g(T)$. Then, $\varepsilon_1(E)$ of Eq. (3) can be used at high temperature as well, by simply replacing $E_0$ and $E_g$ with $E_0[E_g(T)]$ and $E_g[E_g(T)]$, respectively. For the III-V compounds and the structurally similar ternary analogs, the parameter $E_g$ scales linearly with the lowest direct band gap energy $E_g$. As for the $E_d$, it has been shown by Wemple and DiDomenico\(^7\) that $E_d$ is almost independent of $E_g$. Therefore, with the knowledge of the $E_0(E_g)$, and the thermal behaviors of the band gap energy $E_g$, one is able to obtain the refractive index dispersion at a high temperature using the Afromowitz' formula.

### III. CALCULATION AND RESULTS

#### A. AlGaAs materials

For Al$_x$Ga$_{1-x}$As at room temperature, $E_g(x)$, $E_0(E_g)$, and $E_d(x)$ can be expressed as\(^7\)
\[
E_g(x) = 1.424 + 1.266x + 0.26x^2 \quad \text{(eV)};
\]
\[
E_0(E_g) = 2.67 + 0.688E_g;
\]
\[
E_d(x) = 36.1 - 2.45x.
\]
Here the lowest direct band gap energy of Al$_x$Ga$_{1-x}$As is assumed to vary with temperature according to the Varshni's empirical formula, as
\[
E_g(T,x) = E_g(0^\circ \text{C},x) - \frac{\alpha(x) T^2}{T + \beta(x)}.
\]
Since $E_g(298^\circ \text{K},x)$ is known, Eq. (8) can be modified as
\[
E_g(T,x) = \left( E_g(298^\circ \text{K},x) - \frac{\alpha(x) 298^2}{298 + \beta(x)} \right) \frac{\alpha(x) T^2}{T + \beta(x)}.
\]
Although $\alpha(0)$ and $\beta(0)$ have been known well as $5.405 \times 10^{-4}$ eV K$^{-1}$ and 204(K), respectively,\(^11\) the $\alpha(x)$ and $\beta(x)$ for Al$_x$Ga$_{1-x}$As are not known. The values of $\alpha(x)$ and $\beta(x)$ are to be estimated using following information:

(i) $\beta(x)$ is approximately proportional to Debye temperature ($\Theta_D$).\(^12\)

(ii) The Debye temperature, $\Theta_D(x) = 370 + 54x + 72x^2$ (K).\(^13\)

(iii) $E_g(973 \text{ K, } 0.72) = 1.959$ (eV) (He-Ne laser energy).\(^3\)

(iv) $\alpha(0.18) = 6.3 \times 10^{-4}$ eV K$^{-1}$\(^14\). From (i) and (ii), $\beta(x)$ can be directly obtained as
\[
\beta(x) = \frac{204}{370} \left( 370 + 54x + 72x^2 \right) \text{ (K)}.
\]

Note that $\beta(0)$ becomes the value of GaAs. The functional form of $\alpha(x)$ is taken to be a linear function of the alloy composition using available information at $x = 0, 0.18, 0.72,$ as
\[
\frac{\alpha(x)}{\alpha(0)} = \frac{\alpha(0.18)}{\alpha(0)} \times \frac{0.18}{x}.
\]

\[\alpha(x) = 5.405 + 4.038x \times 10^{-4} \text{ eV K}^{-1}.
\]

To obtain this form, $\alpha(0.72)$ was obtained from (iii) and Eq. (9). Then, $\alpha(x)$ was fitted to the linear form, holding $\alpha(0) = 5.405$. Now our model can be applied to AlGaAs materials since the thermal behaviors of the band gap energy and the functional relation of $E_0(E_g)$ are all known. The validity of our model is tested by comparing the experimental data with our calculation. In Fig. 1, one can see the good agreement between calculation and experimental data\(^15\) at 600 °C. The bump near $x = 0.17$ results from the singularity in Eq. (3) at a wavelength of 9400 Å, the band gap of Al$_{0.17}$Ga$_{0.83}$As at 600 °C. The slight deviation for GaAs ($x = 0$) can be explained from the fact that Afromowitz' model itself does not fit well in the region far above band gap. The average deviation is about 0.8% in Fig. 1. Similarly, decent agreement can be seen between the calculated values and the experimental data at 650 °C, at a wavelength of 6328 Å as shown in Fig. 2. The wavelength of 6328 Å is calculated to be the band gap of Al$_{0.69}$Ga$_{0.31}$As at 650 °C, which can be signified also as a bump near $x = 0.69$. The filled triangles in Fig. 2 are our experimental data measured from the laser reflection signals monitored during the metalorganic chemical vapor deposited (MOCVD) growth and the reflection spectroscopy of the grown wafer.\(^16\) The average deviation is about 2% in Fig. 2.
justifiably neglected. Our model could be made more reliable if the dependence of \( E_g \) on \( E_d \) and the information of \( E_g(T,x) \) for \( \text{Al}_{x}\text{Ga}_{1-x}\text{As} \) are fully investigated.

V. CONCLUSION

We propose a scheme to calculate refractive index at high temperature, based on the Afromowitz' formula and the shift of band edge. This idea is applied to AlGaAs materials without using any additional fitting parameter and the calculated results compare quite well with the experimental data. Practically, the results of this work will offer the information on the refractive index at a growth temperature which is necessary for in situ laser reflectometry and other optical monitoring methods for the precision growth of III-V compounds.

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