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## Measurement of Refractive Index of GaP Crystal over a Large Temperature Range Using Interferometry

The refractive index of GaP single crystal was measured through room temperature (300K) to 1200K at a wavelength of 780 nm by using an interferometry with a laser diode. To get a more accurate result, the thermal expansion coefficient of GaP crystal, which would be one parameter for the measurement of the refractive index, was measured by a dilatometer equipped with laser interferometry against temperature in the range from room temperature to 973K. It was confirmed that the linear thermal expansion coefficient was a function of temperature. In this report, an empirical function was obtained to calculate the refractive index at any temperature for GaP crystal. The result shows that the refractive index of GaP varies from 3.1907 to 3.3354 in the temperature range from 300K to 1200K at the wavelength of 780nm.

Keywords: interferometers, thermal expansion, optical constants, semiconductors; non-destructive testing (optical method)

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### 1. Introduction

Refractive index is a most important parameter for a material in the application of optical devices, or for a material being studied via optical method. In the past, many techniques were applied to measure the refractive index of various materials, which were basically stemmed from the following methods: non-destructive interference technique or interferometry [DAMBACH et al., GARCIA QUIRINO et al., GLOBUS et al., KANG et al., MOOSMULLER et al., ROGALA et al., SHEPPARD, TROLINGER], ellipsometry [MALMSTEN et al., KILDEMO et al., WOLFGANG et al., MUNOZ URIBE et al., YUPAPIN et al., ROGALA et al., HODGKINSON et al.], minimum deviation [BOND], Kramers-Kronig Transform from reflectance spectra [PETERSON et al.], X-ray transmission measurements [HARRIS et al.], Z-scan method [EUGENIO et al., MENDONCA et al., ERIKSSON et al.], or some other newly proposed techniques [BEDARIDA et al., TAKEHIRO et al., NIBBERING et al., EL MOSTAFA et al., APOSTOL et al., LIN et al., HUNTINGTON et al.]. As a very important physical parameter, refractive index of GaP has been measured over the past decades by many researchers [ASPNES et al., NELSON et al., KLEINMAN et al., WELKER, PHILIPP et al., PIKHTIN et al.]. However, all the measurements were conducted at room temperature in a range of wavelength. This is not enough when the material is utilized or studied at high temperatures. For example, to get more accurate information about the crystal growth and related phenomena of GaP on a GaP substrate [INATOMI et al., 1997, INATOMI et al. 1993], it is necessary to consider the influence of thermal expansion and refractive index of GaP substrate on the in-situ observation of the crystal growth. To solve this problem, we conducted a measurement of refractive index of GaP crystal from room temperature (300K) to 1200K using an interferometer, which is developed from the in-situ observation system for the crystal growth.

## 2. Experimental

The basic idea of the measurement of refractive index  $n$  comes from the following equation:

$$nd=m\lambda \quad (1)$$

where  $d$  is the thickness of the material,  $m$  is the shifting counts of the interference fringe,  $\lambda$  is the wavelength of the light source. If the crystal is being heated up,  $n$ ,  $d$  and  $m$  will be functions of temperature:  $n(T)$ ,  $d(T)$  and  $m(T)$ . To obtain a reliable function  $n(T)$ ,  $d(T)$  and  $m(T)$  must be accurately determined first.

As a compact and low cost technique, interferometry is a widely used and versatile tool for precision measurement of optical path length ranging from angstroms to tens of meters with high accuracy (better than  $\pm 0.5 \times 10^{-6}$ ) [TROLINGER, BARWOOD et al.]. In this report, we utilized this technique to measure the small increment of the optical path length  $m\lambda$  in GaP substrate caused by thermal expansion.

The actual varying thickness of the crystal  $d(T)$  can be obtained by measuring the thermal expansion coefficient  $\alpha(T)$  of the sample. In the literature [SLACK et al., BERNSTEIN et al., KUDMAN et al., PIERRON et al.], most reports gave one value as the linear thermal expansion coefficient, which were only mean values from the linear approximation over a large temperature range, or only a figure of  $\alpha(T)$  vs. temperature without explicit data or empirical equations. Furthermore, the results were usually contradictory to each other. To get a clearer result, we measured the thermal expansion coefficient of GaP against the temperature over a large temperature range (from 300K to 973K) with a newer setup specially designed to measure thermal expansion and linear thermal expansion coefficient.

The measurement setup for determining the change of the optical path length  $m\lambda$  was developed by an in-situ observation system for observing the solid-liquid interface change during crystal growth [INATOMI et al. 1998]. Fig. 1 shows the setup demonstratively. To obtain fringe pattern in the interferometer microscope, a side of the GaP single crystal slice ( $\phi 7.5\text{mm}$ ) was polished to form a small angle against the other side (also polished to make mirror plane). As GaP single crystal shows low reflectance at the studied wavelength, after polishing, one side of the slice was deposited with a layer of gold (thickness  $\sim 1\mu\text{m}$ ) to increase reflectivity. A gold plate ( $\phi 7.5\text{mm} \times 0.5\text{mm}$ ) was put between the sample slice and a graphite cylindrical tube (size:  $\phi_1=7.5\text{mm}$ ,  $\phi_2=3.5\text{mm}$ ,  $L=25\text{mm}$ ). Both the graphite tube and the gold plate acted as heat flow guide to heat the sample. Gold plate was also used to prevent damaging of the gold film by the thermal couple tip. As gold has very high thermal conductivity, the temperature gradient in the gold plate will be very small, the temperature difference between the measured spot and the sample is thus negligible. The thermal couples were fixed in the middle of the graphite tube by ceramic paste. All these items were installed as shown in Fig. 1 in a quartz glass ampoule (size:  $\phi_1=10\text{mm}$ ,  $\phi_2=8\text{mm}$ ,  $L=235\text{mm}$ ). Then the ampoule was evacuated to a pressure lower than  $10^{-7}$  bar and subsequently sealed completely by melting and merging the quartz opening. The ampoule was finally put into an infrared mirror furnace. The temperature was measured by a thermometer (CHINO KP1000), and the data were transferred to a computer and recorded at the same time. According to the feedback of the temperatures, the computer changed the input power of the halogen lamp to monitor the temperature to the previous heating settings.

An NIR optical system with a laser diode was installed to observe the fringe shifting caused by thermal expansion of the sample through a quartz window. The image beam of the experimental shifting fringe was received in-situ by a CCD camera and then transferred to a videotape through an image processing system.

The angle between the two mirror sides and the thickness of the GaP slice were measured by a confocal laser surface microscope Olympus OLS1000.

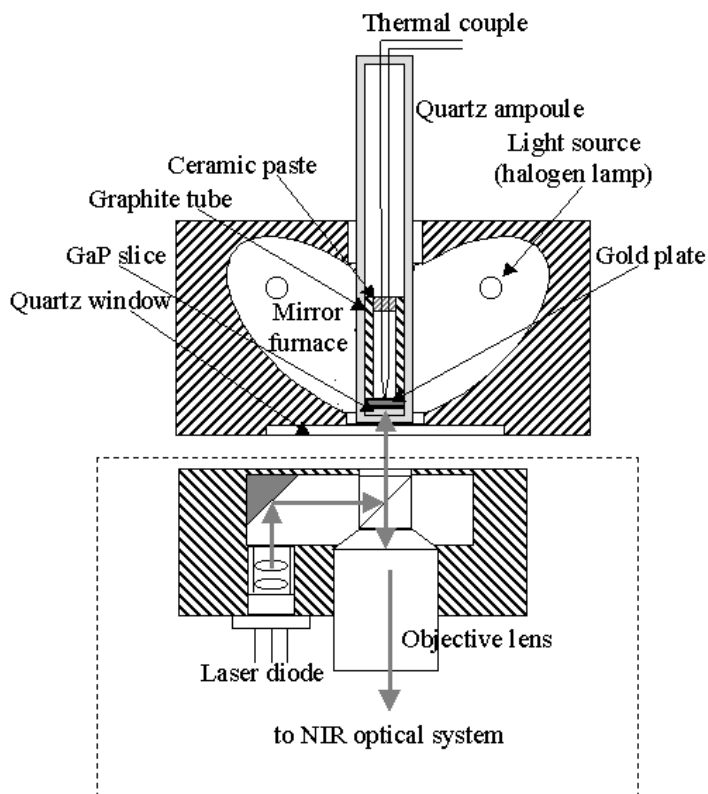


Fig. 1: Schematic setup for measuring high temperature refractive index

The thermal expansion coefficient was measured by a dilatometer (Sinku-Riko LIX-1) equipped with laser interferometry, which is currently the most precise device for measuring thermal expansion with sensitivity up to  $0.02\mu\text{m}$ . The new results were compared with the past work on thermal expansion coefficient of GaP.

### 3. Results and discussion

For the above observation system, we have

$$2\Delta(n \cdot d) = \Delta m \cdot \lambda \quad (2)$$

If the starting temperature is  $T_0$ , the ending temperature is  $T$ , then the refractive index will be  $n(T_0)$  and  $n(T)$ ; the sample thickness will be  $d(T_0)$  and  $d(T)$ ; the shifting counts will be  $m(T_0)$  and  $m(T)$  at the two temperatures, respectively. Thus we get:

$$2n(T)d(T) - 2n(T_0)d(T_0) = [m(T) - m(T_0)]\lambda \quad (3)$$

From this equation, the refractive index of the sample at any temperature  $T$  can be obtained:

$$n(T) = \frac{[m(T) - m(T_0)]\lambda + 2n(T_0)d(T_0)}{2d(T)} \quad (4)$$

where

$$d(T) = d(T_0) + d(T_0) \int_{T_0}^T \alpha(T) dT \quad (5)$$

$\alpha(T)$  is the linear thermal expansion coefficient of the sample at temperature  $T$ .

Table 1 lists the preparation conditions of the sample for the measurement of the shifting counts during the heating process.

Orientation	GaP (111)
Thickness at the center	322.2 $\mu\text{m}$
Angle between the two sides	$10.27 \times 10^{-3}$ (rad)

Table 1: Preparation conditions of GaP slices for the measurement of the shifting counts

Fig. 2 shows the variation of shifting counts with the temperature. It can be seen that the shifting counts increased with increasing temperature. By using the least square method, the curve was fitted with a polynomial expression and a function was obtained as follows:

$$m(T) = -29.2551 + 0.095313T + 9.0003 \times 10^{-6} T^2 + 2.06945 \times 10^{-8} T^3 \quad (6)$$

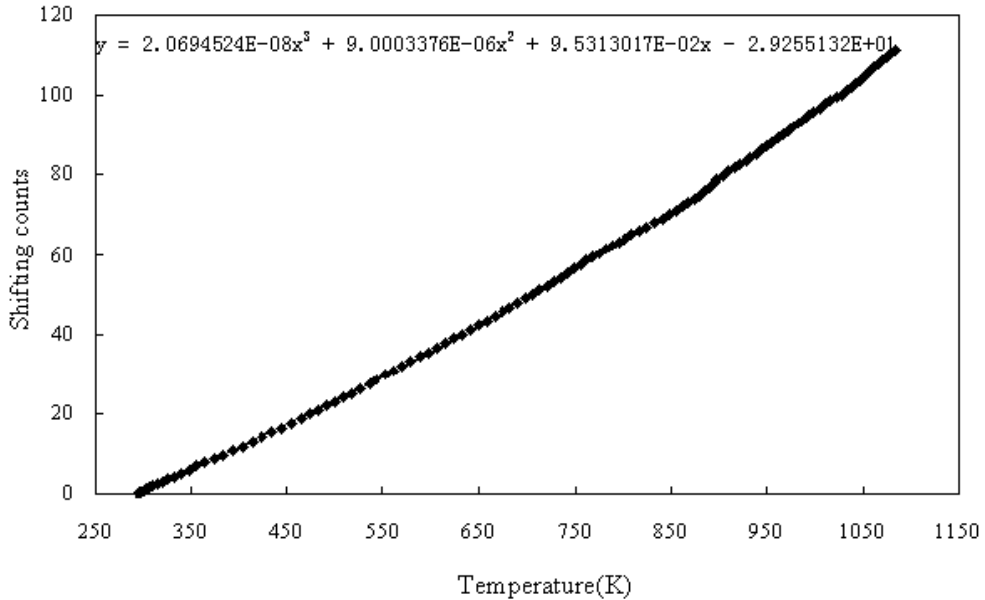


Fig. 2: Variation of shifting counts with temperature

Fig. 3 shows the experimental results for the measurement of the linear thermal expansion coefficient of a sample (orientation (100), thickness 0.305mm) and the fitted curve obtained by the following equation from a semi-empirical quasi-harmonic model [REEBER et al.]:

$$\alpha(T) = \sum_{i=1}^n X_i \frac{(\theta_i / T)^2 \exp(\theta_i / T)}{[\exp(\theta_i / T) - 1]^2} \quad (7)$$

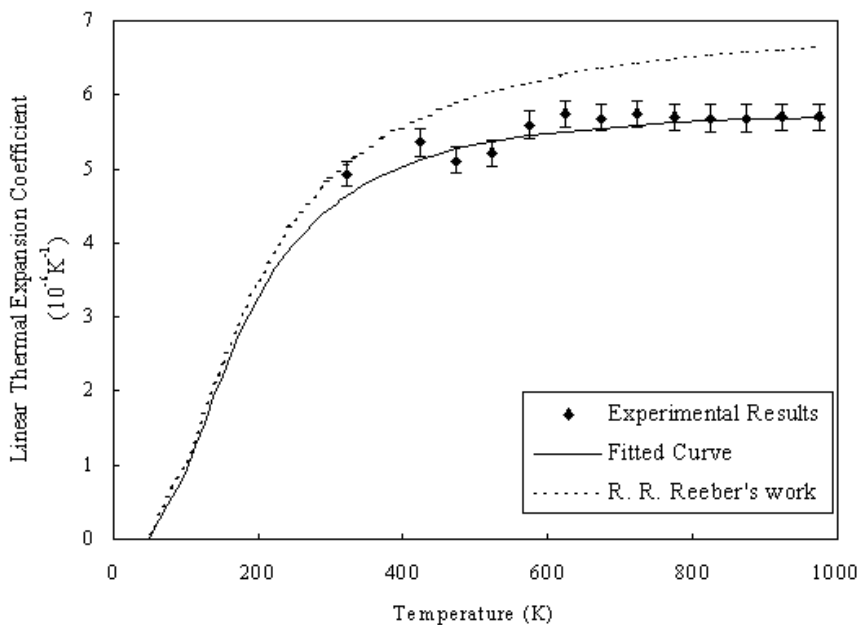


Fig. 3: Experimental results and curve fitting for the measurement of the linear thermal expansion coefficient of a sample (orientation (100), thickness 0.305mm)

The fitting parameters are listed in Table 2:

$\theta_1(K)$	$\theta_2(K)$	$\theta_3(K)$	$\theta_4(K)$	$X_1(10^{-7}/K)$	$X_2(10^{-7}/K)$	$X_3(10^{-7}/K)$	$X_4(10^{-7}/K)$
30	200	600	1552.5	-3.2721	11.1199	51.8202	-1.1249

Table 2: Fitting parameters for the linear thermal expansion coefficient of GaP

From Fig. 3, it can be seen that the linear thermal expansion coefficient of GaP is a function of the temperature. As a comparison, R. R. Reeber’s work was presented in this figure. It is clearly seen that the new result is similar to Reeber’s work. However, the experimental data is a little smaller than the latter. In the present work, the error is  $\pm 0.18 \times 10^{-6}$ .

From equation (4), (5) and (7), we can obtain the expression of the refractive index:

$$n(T) = \frac{[m(T) - m(T_0)]\lambda + 2n(T_0)d(T_0)}{2d(T_0) \left\{ 1 + \sum_{i=1}^4 \left[ \frac{X_i \theta_i}{\exp(\theta_i / T) - 1} \right] \Big|_{T_0}^T \right\}} \tag{8}$$

Refractive index at room temperature 300K at the wavelength 780nm can be obtained from W. L. Bond’s work [BOND]:  $n(300K) = 3.1907$

If  $T_0 = 300K$ , then

$$n(T) = \frac{3.154432 + 1.153696 \times 10^{-4} T + 1.089422 \times 10^{-8} T^2 + 2.504921 \times 10^{-11} T^3}{\sum_{i=1}^4 \left[ \frac{X_i \theta_i}{\exp(\theta_i / T) - 1} \right] + 0.999373} \tag{9}$$

This is the final expression of refractive index of GaP as a function of temperature at the wavelength 780 nm.  $X_i$  and  $\theta_i$  can be found in Table 2. From this equation, we can obtain the refractive index at any temperature.

Fig. 4 shows the final result for the relationship between the refractive index of GaP and temperature at the wavelength 780nm. From the figure, we can see that the refractive index increases almost linearly with increasing temperature. To get a clearer overview, we calculated the indices at different temperatures and listed the results in Table 3. From the table, it can be seen that as the temperature increases from 300K to 1200K, the refractive index increases from 3.1907 to 3.3354.

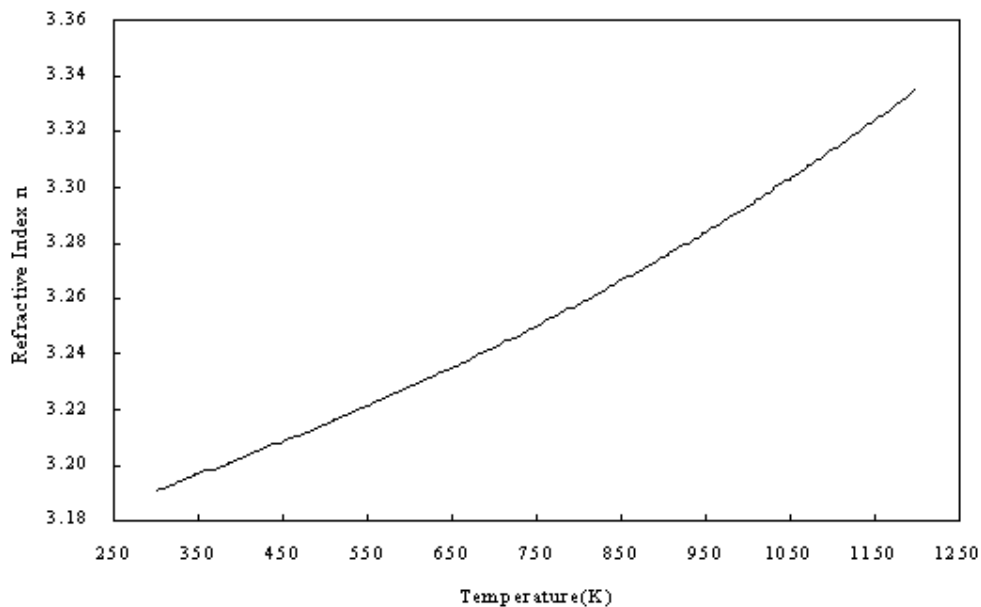


Fig. 4: The relationship between the refractive index of GaP and temperature at the wavelength 780nm

$T$ (K)	$n(T)$ ( $\pm 0.0128$ )
300	3.1907
350	3.1965
400	3.2024
450	3.2085
500	3.2148
550	3.2213
600	3.2280
650	3.2350
700	3.2423
750	3.2499
800	3.2579
850	3.2662
900	3.2748
950	3.2838
1000	3.2933

1050	3.3031
1100	3.3134
1150	3.3242
1200	3.3354

Table 3: Refractive indices of GaP at different temperatures at the wavelength 780 nm

As the measurement of the linear thermal expansion coefficient was undertaken from the room temperature to 700°C, the above results in the temperature range 300~973K were more reliable. However, as the semi-empirical quasi-harmonic model can be safely extrapolated to higher temperatures [REEBER et al.], the calculated refractive index from function (9) should also be reliable.

#### 4. Conclusion

In conclusion, the refractive index of GaP single crystal was measured over a large temperature range. An empirical equation of the index as a function of temperature was obtained to calculate the index at the wavelength 780nm for GaP single crystal. The linear thermal expansion coefficient was also measured, and an equation was obtained by using QHM model. The result showed that the refractive index of GaP increases from 3.1907 at 300K to 3.3354 at 1200K.

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