

Determination of Optical Constant of Materials by two Different Methods: An Application to Single Crystals Si

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Received 02.02.1999

Abstract

The normal reflectance of single crystal Si was measured at 0.5 5.6 eV range by unpolarized light. Optical parameters of Si were determined from this reflectance data by using two different methods: oscillator fit procedure and Kramers-Kronig analysis. The refractive index, extinction coefficient and the real and imaginary parts of complex dielectric constant obtained by these two methods were compared. A good agreement was found between two methods. Furthermore, the optical parameters so obtained seem to be in a satisfactory agreement with the literature.

Key Words: Optical Constant, Si Single Crystals, Oscillator Fit Procedure, Kramers-Kronig Analysis.

1. Introduction

There are many different methods that have been developed for the purpose of determining the optical constants of materials. Some of these are based upon measurement of multiple reflections or transmission and absorbance in thin films, while others are based on the measurement of reflectance from bulk materials only. Every method has differences with respect to their relative precision and the techniques with which they handle the experimental data. Depending on the experimental setup, the optical constants have been determined either only at the specified energy values or over some energy range.

Most common techniques that have been used to determine the optical constants over the whole measurement range are Kramers-Kronig analysis [1,2] and classical oscillator fit procedure [3]. The former, in its most commonly used form, is based on the measurement

of normal reflectance, over an energy range, from a bulk material and the application of an integral transformation. If the material considered is in the form of a thin film or is transparent, the application of this method becomes difficult, since no simple or clear expression can be written for this case. The most important disadvantage of this method is the probable errors arising from taking into account the contributions from outside the experimental measurement range by means of extrapolation.

In the other commonly used technique, the optical parameters of investigated materials are represented as a sum of classical oscillators in the related spectral region. In this method, the complex dielectric constant is generally, written as a sum of damped classical oscillators. These oscillators' parameters are determined by fitting the chosen dielectric function to experimental data. The most important disadvantage of this method is that the fit procedure requires long calculations. Especially, if the experimental reflectance spectra show marked structure, too many oscillators have to be chosen to provide a satisfactory fit.

2. Theory

It is well known from Fresnel equation that, in any solid material, the complex reflectance is given as

$$\tilde{r} = \sqrt{R(\omega)}e^{i\theta(\omega)}. \quad (1)$$

Here, the $R(\omega)$ is measured reflectance, and $\theta(\omega)$ is phase shift arising from the reflection. By taking logarithm of Eq. (1), one obtains

$$\ell n(\tilde{r}) = \ell n[\sqrt{R(\omega)}] + i\theta(\omega). \quad (2)$$

If Hilbert transformation is applied to this expression, the real and imaginary parts of it can be related to each other as

$$\theta(\omega) = -\left(\frac{\omega}{\pi}\right)P \int_0^{\infty} \frac{\ell n(R(\omega'))}{\omega'^2 - \omega^2} d\omega' \quad (3)$$

Here, P is the principal value of the integral.

In the process of performing the Kramers-Kronig analysis, Eq. (3) is first applied to the measured normal $R(\omega)$ data taken between a certain $\omega_a - \omega_b$ frequency range and $\theta(\omega)$ are calculated for each value. The most important problem encountered here is to calculate the contributions arising from outside the measured range, namely $(0 - \omega_a)$ and $(\omega_b - \infty)$. The integral corresponding to the measured range can be calculated by means of any numerical integration technique. For the purpose of accounting for the contributions from outside of the range, various authors have supposed different extrapolation functions [4, 5]. The most common and the oldest of these is Roessler's extrapolation function, in which the contributions from the outside are represented by a logarithmic peak having its maximum value at the end points. In this study, this extrapolation function has been used [4].

For solid materials, if $R(\omega)$ and $\theta(\omega)$ are known, the refractive index $n(\omega)$ and extinction coefficient $k(\omega)$ of that material are given by

$$n(\omega) = \frac{1 - R(\omega)}{1 + R(\omega) - 2\sqrt{R(\omega)} \cos \theta(\omega)}, \quad (4)$$

$$k(\omega) = \frac{-2\sqrt{R(\omega)} \sin \theta(\omega)}{1 + R(\omega) - 2\sqrt{R(\omega)} \cos \theta(\omega)}. \quad (5)$$

Moreover, the complex dielectric constant of a solid is given as

$$\tilde{\epsilon}(\omega) = \epsilon_1(\omega) + i\epsilon_2(\omega). \quad (6)$$

Here, real and imaginary parts are related to $n(\omega)$ and $k(\omega)$ as

$$\begin{aligned} \epsilon_1(\omega) &= n^2(\omega) - k^2(\omega), \\ \epsilon_2(\omega) &= 2n(\omega)k(\omega). \end{aligned} \quad (7)$$

Another method that is used widely to determine the optical constant is the oscillator fit procedure. In the spectral range involving the lattice vibrations, the complex dielectric constant of a solid is represented as a sum of classical oscillators as

$$\tilde{\epsilon}(\omega) = \epsilon_\infty + \sum_i^N \left[\frac{S_i}{1 - (\frac{\omega^2}{\omega_i^2}) - iF_i(\frac{\omega}{\omega_i})} \right] \quad (8)$$

Here, S_i , ω_i and F_i are oscillator strength, frequency, and bandwidth of the i 'th oscillator respectively [3]. ϵ_∞ is the contribution to the dielectric constant due to high frequencies, especially from electronic transitions, and is called the "**high frequency dielectric constant**".

On the other hand, it is known [6] that, for normal incidence, the reflectance of any material is related to the dielectric constant as,

$$R(\omega) = \left| \frac{\sqrt{\tilde{\epsilon}(\omega)} - 1}{\sqrt{\tilde{\epsilon}(\omega)} + 1} \right|^2 \quad (9)$$

The real and imaginary parts of the complex dielectric constant can be given by

$$\epsilon_1(\omega) = n^2(\omega) - k^2(\omega) = \epsilon_\infty + \sum_i^N \left[\frac{S_i(1 - \frac{\omega^2}{\omega_i^2})}{(1 - (\frac{\omega^2}{\omega_i^2})) + F_i^2(\frac{\omega}{\omega_i})^2} \right], \quad (10)$$

$$\epsilon_2(\omega) = 2n(\omega)k(\omega) = \sum_i^N \left[\frac{S_i F_i \frac{\omega}{\omega_i}}{(1 - (\frac{\omega^2}{\omega_i^2}))^2 + F_i^2(\frac{\omega}{\omega_i})^2} \right]. \quad (11)$$

On the other hand, the complex dielectric constant can be written in terms of reflectance and phase angle [7]:

$$\tilde{\epsilon}(\omega) \left(\frac{1 + \sqrt{R(\omega)}e^{-i\theta(\omega)}}{1 - \sqrt{R(\omega)}e^{-i\theta(\omega)}} \right)^2. \quad (12)$$

If the $\theta(\omega)$ phase angle is withdrawn from this expression, following equation can be written as

$$\theta(\omega) = -\arcsin \left(\frac{(R(\omega) - 1) \left(\frac{\epsilon_1}{\epsilon_2} - \sqrt{\left(\frac{\epsilon_1}{\epsilon_2} \right)^2 + 1} \right)}{2\sqrt{R(\omega)}} \right), \quad (13)$$

which is an analytical expression depending on the real and the imaginary parts of dielectric constant and on the reflectance.

3. Experimental Procedure

The normal reflectance measurements of single crystal Si wafers was performed by a JASCO model UV-VIS-IR spectrophotometer, in the 220-2200 nm wavelength range. Prior to the measurement, the samples were left in a HF-H₂O mixture for adequate time to remove a possible oxide layer. Reflectance measurements have been made at room temperature and by using unpolarized light. The data taken in two nanometers intervals are transferred into a computer. For the Kramers- Kronig Analysis, a Fortran program has been written, and the numerical integration has been performed by Simpson 1/3 integration method. Moreover, in the oscillator fit procedure, Sigma-plot 1.0 program has been used.

4. Results and Discussion

Oscillator Fit Method.

The experimental reflectance spectrum is given in Fig. 1. As seen from the figure, there is a uniform rise until 3.5 eV. Two main peaks, $E_1 = 3.4$ eV and $E_2 = 4.49$ eV, correspond to high energy interband transitions. Vina and Cardona [8] have reported similar energy values for these transitions such as $E_1 = 3.4$ eV and $E_2 = 4.25$ eV. They have attributed E_1 energy to the interband transition from highest valance band to lowest conduction band along the Λ symmetry line in the Brillouin zone [8]. In fact, this peak has represented the direct transitions occuring at the L and Γ points. In the analysis of Germanium's reflectance spectrum (which is similar to that of Si), these transitions are distinguishable from each other, while in Si's reflectance, these two transitions have the same energy, and therefore seem to be a single peak. The E_2 peak arises from wide regions of Brillouin zone, especially near a special $\frac{2\pi}{a}(\frac{3}{4}, \frac{1}{4}, \frac{1}{4})$ point [8,9].

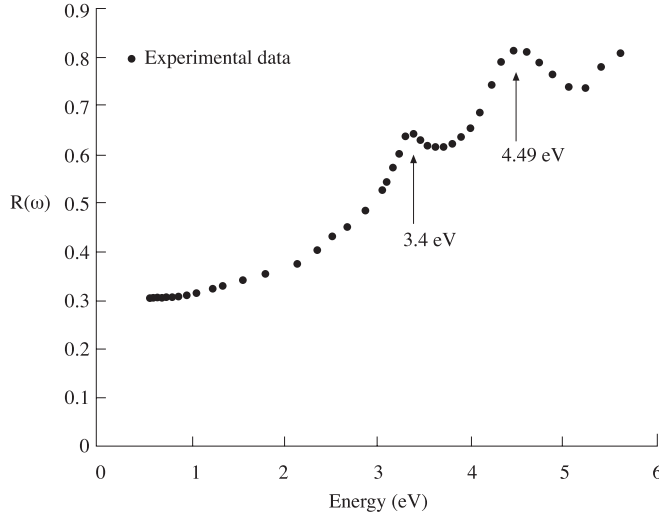


Figure 1. Experimental reflectance spectrum of Si.

One of the most important problems encountered in classical oscillator fit procedure is to decide how many oscillators be chosen within the experimental spectral range, and whether to take into account any contribution from outer parts of related regions. Generally the number of oscillators required can be determined from the reflectance spectrum considered. But, in performing the fit procedure, it is assumed that, in addition to the oscillators selected in the experimental reflectance spectrum, it can be useful (for better fit) to choose more oscillators outside of this range [6]. But in this work, the oscillators selected within the measurement range had provided a satisfactory result and therefore any oscillators outside had not been selected. The number and positions of oscillators have been determined by means of derivative of experimental reflectance spectrum. In this manner, six oscillators have been chosen and these initial oscillator parameters and their values found by fit procedure have been given in Table 1. In Fig. 2, the reflectance curve plotted by using the initial oscillator parameters and the experimental curve are compared. In Fig. 3, the reflectance curve obtained by using the final values of oscillator parameters and experimental curves have been given for comparison.

As seen from Figure 3, there is a perfect agreement between the experimental and theoretical curve, which is plotted by the parameters, found in the fit procedure. To prevent the time loss and to speed the procedure, not all of the measurement values but only some selected values which represent the whole spectral variation have been employed. By using the final oscillator parameters in Eq. (9) and (10), $\epsilon_1(\omega)$ and $\epsilon_2(\omega)$, real and imaginary parts of complex dielectric constant have been calculated and given in Figure 4. Phase shifts calculated by Eq.(12) is shown in Figure 5. On the other hand, by making use of these phase values and also reflectance values in Equations 4 and 5, the refractive index $n(\omega)$ and the extinction coefficient $k(\omega)$ are determined and given in Figure 6.

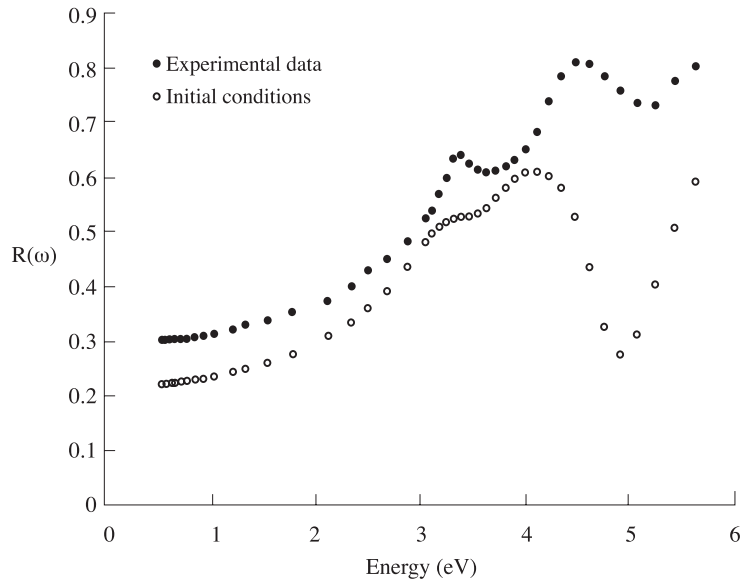


Figure 2. Theoretical reflectance curve obtained by initial oscillator parameters and the experimental one.

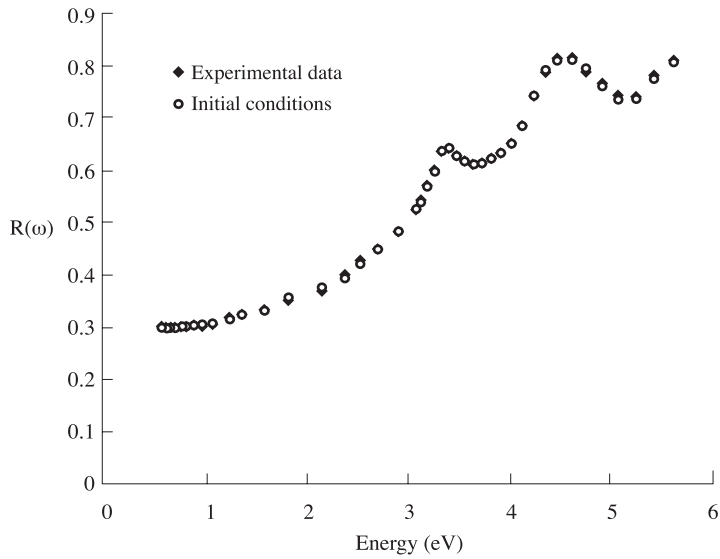


Figure 3. Comparison of experimental and theoretical reflectance spectra which is obtained after fit.

Table 1. The initial and final values of oscillator parameters (Initial $\epsilon_\infty = 2$, after the fit $\epsilon_\infty = 0.787$).

S_i		ω_i		F_i	
Initial	From the fit	Initial	From the fit	Initial	From the fit
1.8	0.8102	3.2	3.4848	0.15	0.0657
1.8	1.6044	3.4	3.3647	0.15	0.0503
1.8	3.3142	3.7	3.6555	0.15	0.1619
1.8	4.8913	3.9	4.0090	0.15	0.1771
0.5	2.7924	4.2	4.2375	0.15	0.0837
1.8	1.2181	5.4	5.2424	0.15	0.1664

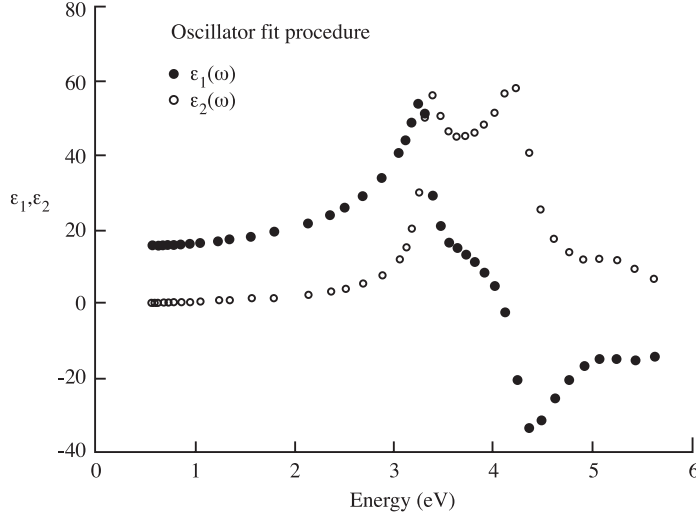


Figure 4. Real and imaginary parts of complex dielectric constant of Si obtained by oscillator fit procedure.

Kramers-Kronig Method.

As a second method in this work, Kramer-Kronig analysis has been applied. For this purpose, by using the reflectance values and Eq. (3), corresponding phase shifts are calculated and shown in Figure 7. In calculating these phases, the Roessler’s extrapolation function has been used. In substituting these phase values and experimental reflectance data in Eq. (4) and (5), $n(\omega)$ and $k(\omega)$ are calculated and from equation 6, $\epsilon_1(\omega)$ and $\epsilon_2(\omega)$ are determined. and are shown in Figure 8, and $\epsilon_1(\omega)$ and $\epsilon_2(\omega)$ in Figure 9.

In Roessler’s method, to take into account the contributions from the outside of the measurement range, two wavelengths have been selected over the measurement range at which total phase shift are assumed to be zero. Many investigators have supposed that the parameters to be determined are very sensitive to this selected wavelength pair.

It is declared that this wavelength pair has need not be near the end of experimental measurement range.

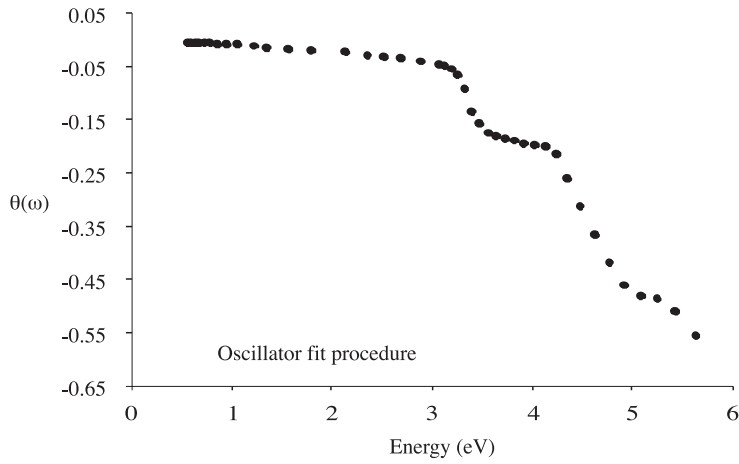


Figure 5. Phase shifts upon reflection calculated by using Equation 12.

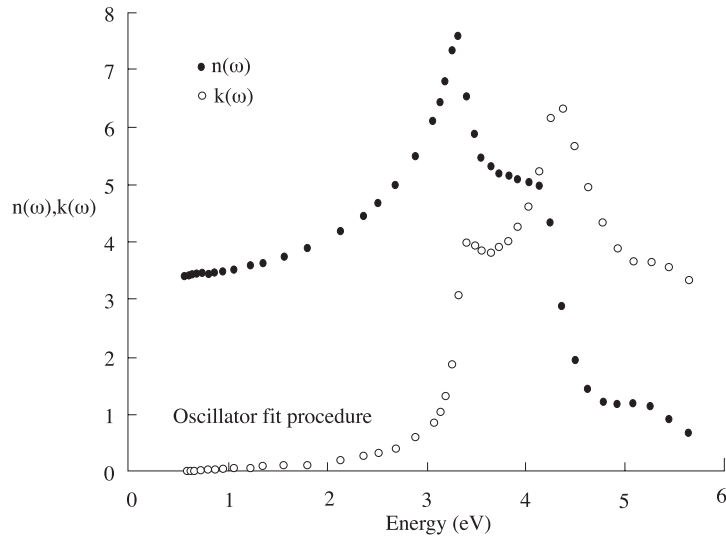


Figure 6. Refractive index and extinction coefficients calculated by means of Equation 4 and 5.

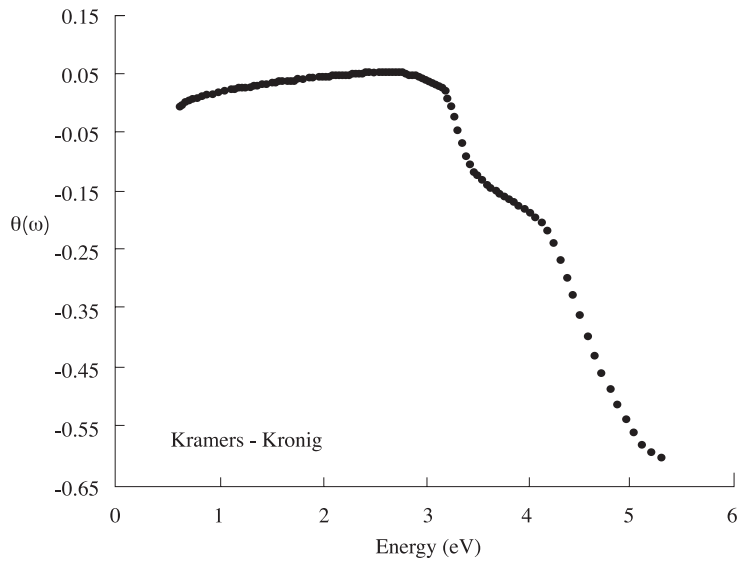


Figure 7. Phase shifts variation determined by Kramers-Kronig transformation.

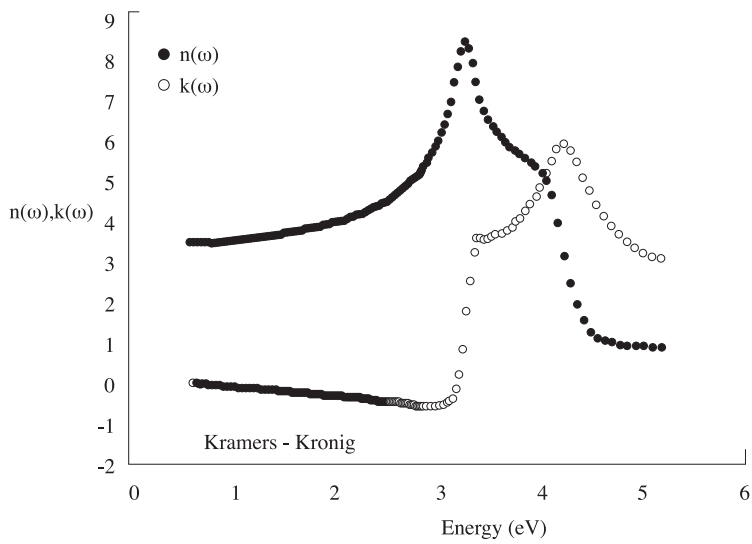


Figure 8. Refractive index and extinction coefficient determined by KKT.

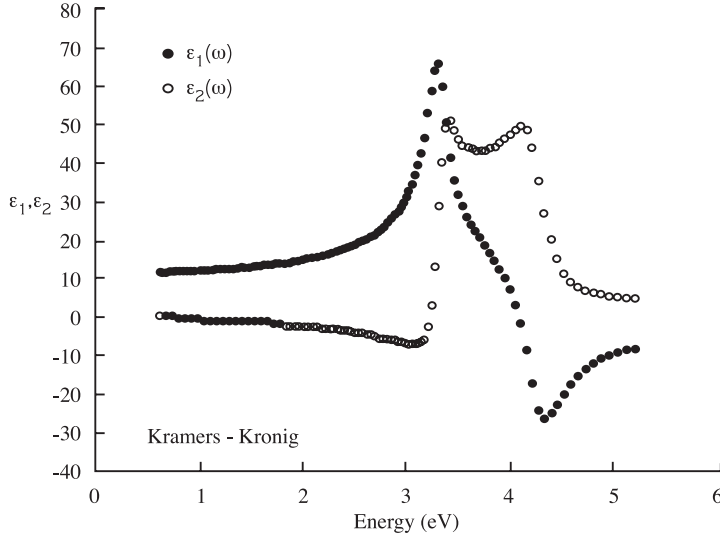


Figure 9. Real and imaginary parts of complex dielectric constant determined by KKT.

In this work, the 1602 and 1632 nm wavelength pair are chosen. These values correspond to lower energies than forbidden energy gap of Si (1.1 eV). Two phase shifts, that are obtained by this wavelength pair and by oscillator fit procedure, have been found very close to each other. Apart from this mentioned pair, a number of various wavelength pairs have been selected and it was seen that refractive index, $n(\omega)$ is not too dependent on this pair selection. But, $k(\omega)$ extinction coefficient and hence imaginary part of complex dielectric constant, $\epsilon_2(\omega)$, are found to be more sensitive to the selection of wavelength pair.

If Fig. 6 and 8 are compared, it can be seen that the refractive indices obtained by two methods show almost the same variation but extinction coefficients exhibit some difference. This discrepancy is due to the difference between the phases obtained by the two methods, especially at higher angles. The $n(\omega)$ values obtained by the two methods, have been found to be in good agreement with the literature [10].

By considering Figs. 4 and 9, it can be said that the above mentioned ideas for $n(\omega)$ and $k(\omega)$ are also true both for $\epsilon_1(\omega)$ and $\epsilon_2(\omega)$. $\epsilon_1(\omega)$ curves determined by two methods are in a good agreement with each other [8, 9]. There are two main peaks in $\epsilon_2(\omega)$ graph. These correspond to main peaks in the reflectance spectrum, having energies of $E_1 = 3.4$ eV and $E_2 = 4.49$ eV. The positions of these peaks are consistent with the literature [8].

5. Conclusions

KKT analysis is still most commonly used technique in evaluating the experimental reflectance measurements. Although two different methods used here, KKT analysis and oscillator fit procedure, are independent in principle, they might be used as complemen-

tary tools for each other.

There are various approximations for phase correction in KKT [11, 12]. The KKT method could be applied for both bulk materials and thin films and also for anisotropic media [13]. But, as is well-known, this analysis is very sensitive to the method of extrapolation. Therefore, the results obtained by it must be tested by other methods. Here, this comparison was performed for Si and it is found that the results obtained by KKT and fit procedure are in agreement with each other.

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