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Optical properties of Zinc Selenide slabs at 373 and 423 K in the wavelength 2-15 μ m

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1 Optical properties of Zinc Selenide slabs at 373 and 423 K in the wavelength 2-15 μ m
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8 **ABSTRACT** Zinc selenide (ZnSe) is a nonlinear optical crystal and infrared
9 optical material, and its window exhibits unique optical properties in the various
10 optical and electronic devices applications. Transmittance spectrum of ZnSe slabs in
11 the infrared wavelength 2-15 μ m at normal incidence were investigated by a Bruker
12 V70 FTIR spectrometer. The optical constants of ZnSe at 373 and 423K were
13 achieved by modeling the transmittance spectrum of ZnSe slab. Then the optical
14 properties of single and double slabs were obtained, including transmittance,
15 reflectance and absorptance. The results show that the shapes of transmittance,
16 reflectance and absorptance spectrum of ZnSe slabs at 373 and 423K are similar, and
17 single and double ZnSe slabs all have a wide transparent region. With the temperature
18 increasing, the extinction coefficient of ZnSe decreases, but the refractive index of
19 ZnSe increases. The thicker the thickness of ZnSe slab, the weaker the transmittance
20 and reflectance of ZnSe slab, and the stronger the absorptance of ZnSe slab.

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Key words: Zinc Selenide; optical properties; transmittance spectra

1. Introduction

Zinc selenide (ZnSe) is an important and promising, direct band gap compound semiconducting material for applications to the various optical and electronic devices [1-5], such as blue-green laser diodes (LDs), white light emitting diodes (LEDs), optically controlled switches, tunable mid-IR laser sources for remote sensing applications, photovoltaic and photo electro-chemical devices. ZnSe is also used as a window in solar cells together with some narrowband gap semiconductor[6, 7]. Optical properties of ZnSe are the basic optical parameters for analyzing ray transfer in ZnSe window [27], and measuring optical constants of liquid sample in optical cell that is consisted of double ZnSe slabs[8-10, 28].

The researchers have been attracted by the optical properties of ZnSe for more than 50 years [11-17]. Hattori et al. [11] measured the refractive index of ZnSe in the spectral region from 10 to 100 cm^{-1} . Ahmed et al. [12] experimentally investigated the refractive index of ZnSe in the spectral region from 2.5 to 14 μm . Jensen et al. [13] obtained the refractive index of ZnSe from the quantum mechanical calculation of the dielectric constant of a compound semiconductor. Li [14] reviewed early researches about optical properties of ZnSe, and reported 36 sets of experimental data available for the refractive index of ZnSe. Palik [15] also presented a review of early research works published before 1990s, in which a limited optical constants of ZnSe were also presented. Recently, Li et al. [16, 17] measured transmittance spectrogram of ZnSe glass in the infrared wavelength 0.83–21 μm at normal incidence by Bruker V70

FTIR spectrometer, and investigated the optical constants of ZnSe glass. However, the optical properties of ZnSe published in the early times were limited in the wavelength region, and most researches did not analyze the temperature effect on the optical properties of ZnSe.

The optical properties of ZnSe slab can be calculated by the optical constants of ZnSe, which include the extinction coefficients and refractive index. The present measurement techniques of optical constants of solid materials are outlined as following: inversion methods based on reflectance and transmittance spectrograms [18-21], single reflectance method [22] and single transmittance method [23-24], spectroscopic ellipsometry [25-26], and double-thickness transmittance method [16-17]. El-Zaiat et al. [21] measured spectral reflectance an incidence angle of 5 to the normal to external slab faces and transmittance at normal incidence of the glass sample, and developed an inversion method based on reflectance and transmittance spectrograms to calculate the real and imaginary parts of the refractive indices. The inverse calculation process of the single reflectance and the single transmittance methods need to combine with Kramers-Kronig ($K-K$) dispersion equation [22-24]. The mechanism of double-thickness transmittance method is simple, and $K-K$ equation is not introduced in the inverse calculation [16, 17].

In this work, a method to determine optical constants of ZnSe with two transmittance spectrum of ZnSe slab was introduced in the reference [16]. The method was validated by experimentally determining optical properties of ZnSe at room temperature. Then measurements of transmittance spectrogram of ZnSe slab in the

infrared wavelength 2-15 μm were investigated by a Bruker V70 FTIR spectrometer.

The optical properties of single and double ZnSe slabs at 373 and 423K were achieved by the experimental method.

2. Experimental Method and Validation

Spectrophotometric measurements at normal incidence were made using a Bruker V70 FTIR spectrometer with a wavelength range 2-15 μm . Light from the FTIR's internal light source was focused through an iris, collimated, and passed into a Michelson interferometer that caused each wavelength to be modulated. This modulated beam was then passed through the ZnSe slab and focused onto an HgCdTe or RT-DLaTGS detector. The detector signal was analyzed with the manufacture's software (OUPS version 6.5), which uses Mertz phase correction and boxcar apodization without zero filling to provide spectrally resolved transmitted intensity.

For each measurement, the transmittance is as a function of the transmitted and incident intensity of light as follows:

$$T = \frac{I}{I_0} \quad (1)$$

Where, I_0 and I are the incident (baseline) and transmitted intensities of light, respectively. The incident intensity was measured with the ZnSe slab removed from the beam's path. The transmitted intensity was measured with the ZnSe slab placed in the optical path. Measurements were made with a resolution of 4 cm^{-1} . As shown in Fig.1 [28], the ZnSe slab is fixed in an oven along with an temperature controller allowed the temperature of the cell to be controlled to within 5 $^{\circ}\text{C}$.

The reflectance, transmittance and absorptance of a single ZnSe slab at normal incidence are given as [16]

$$R_s = \rho + \frac{(1-\rho)^2 \rho \exp(\frac{-8\pi kL}{\lambda})}{1-\rho^2 \exp(\frac{-8\pi kL}{\lambda})} \quad (2a)$$

$$T_s = \frac{(1-\rho)^2 \exp(\frac{-4\pi kL}{\lambda})}{1-\rho^2 \exp(\frac{-8\pi kL}{\lambda})} \quad (2b)$$

$$A_s = 1 - T - R \quad (2c)$$

Where, R_s , T_s and A_s are the reflectance, transmittance and absorptance of a single ZnSe slab at normal incidence, respectively. ρ is the interface reflectance for surface between air and slab. k is the extinction coefficient of ZnSe material. L is the thickness of single ZnSe slab. λ is the wavelength.

The interface reflectance ρ are calculated based on Fresnel's relations.

$$\rho = \frac{(n-1)^2 + k^2}{(n+1)^2 + k^2} \quad (3)$$

Where, n is the refractive index of ZnSe material.

The reflectance, transmittance and absorptance of double spliced ZnSe slabs at normal incidence, in which exists air aperture between the slabs, are given as[17]

$$R_d = R_s(1 + \frac{T_s^2}{1-R_s^2}) \quad (4a)$$

$$T_d = \frac{T_s^2}{1-R_s^2} \quad (4b)$$

$$A_d = 1 - R_d - T_d \quad (4c)$$

Where, R_d , T_d and A_d are the reflectance, transmittance and absorptance of double ZnSe slabs at normal incidence, respectively.

For establishing the reflectance and absorptance of the single and double ZnSe slabs with Eqs. (2) and (4), the optical constants of ZnSe material are firstly required.

If the extinction coefficient and refractive index of ZnSe material in the wavelength λ are known, the measured transmittances T_1 and T_2 of the single ZnSe slab at normal incidence with the slab thickness L_1 and L_2 can be given by[16]

$$T_1 = \frac{(1-\rho)^2 \exp(\frac{-4\pi k L_1}{\lambda})}{1-\rho^2 \exp(\frac{-8\pi k L_1}{\lambda})} \quad (5a)$$

$$T_2 = \frac{(1-\rho)^2 \exp(\frac{-4\pi k L_2}{\lambda})}{1-\rho^2 \exp(\frac{-8\pi k L_2}{\lambda})} \quad (5b)$$

A double-thickness transmittance method was introduced in the reference[16]. The relationship equations obtained from Eqs. (5a) and (5b) are presented as follows[16]

$$\rho = \frac{1 - \sqrt{T_1^2 + T_1 [\exp(4\pi k L_1 / \lambda) - \exp(-4\pi k L_1 / \lambda)]}}{1 + T_1 \exp(-4\pi k L_1 / \lambda)} \quad (6)$$

$$k = \frac{\lambda}{4\pi L_2} \ln \left[(1 + \sqrt{1 + 4c^2 \rho^2}) / (2c) \right] \quad (7a)$$

$$c = \frac{T_2}{(1-\rho)^2} \quad (7b)$$

From Eq. (3), refractive index of ZnSe material n can be calculated by

$$n = \frac{(1+\rho) + \sqrt{(1+\rho)^2 - (1-\rho)^2(1+k^2)}}{1-\rho} \quad (8)$$

To solve the above analytical expressions, a calculation procedure based on iteration technique was adopted as the following steps: 1) give the assumed k values; 2) calculate the ρ values by Eq. (6), and determine the new k values by Eq. (7); 3) analyze the calculation errors between the assumed k values and the new k values. If the errors are rational, the k values calculation is finished, otherwise the assumed k values will be substituted by the new k values and then return to 2); 4) calculate n

values by Eq. (8) on the basis of k values.

To validate our experimental and calculation procedure, we compared our measurements of ZnSe with published data. The experimental transmittance spectrum of the single ZnSe slabs in wavelength 2-15 μm at room temperature, whose layer thickness is respectively 3 and 4mm, are shown in Fig. 2. The optical constants of ZnSe were established by the method of present work. The experimental results of this work and public data in the reference [14,15] are together shown in Fig.3 and Fig.4. As shown in Fig.3, most extinction coefficients of ZnSe are in agreement with the published data. However, from Fig.4 the refractive indexes of this work in the wavelength 2-15 μm are lower than ones in Ref. 14, but the relative errors of the refractive indexes between this work and Ref. 14 are less than 10%. The reason is that the optical constants in Ref. 14 were obtained between 1960s and 1980s, when more attention was paid to refractive index of ZnSe, whereas the extinction coefficients of ZnSe were omitted.

3. Result and Discussions

3.1 Experimental Measurements

The experimental transmittance spectrum of ZnSe slabs at 373 and 423K in the wavelength 2-15 μm , whose layer thickness is 3 and 4mm respectively, are shown in Fig. 5.

As shown in Fig.5, it is clear that the shapes of transmittance spectrum between 3 and 4 mm ZnSe slab at 373 and 423K are similar, and the these ZnSe slabs all have a wide transparent region, in which the transmittances of ZnSe slabs are beyond 66%.

However, the optical transmittance performance of ZnSe slabs at 373 and 423K is much weaker than at room temperature in the Fig.2, and with the temperature of ZnSe slabs increasing, the optical transmittance performance of ZnSe slabs decreases, which shows that the temperature plays an important role in the optical properties of ZnSe slabs.

3.2 Determination of Optical Constants

The optical constants n and k of ZnSe at 373 and 423K in the wavelength region 2-15 μ m are attained by the experimental method in present work based on experimental values from Fig.5. The extinction coefficients and refractive indexes of ZnSe are plotted in Fig. 6 and Fig. 7.

Fig.6 shows the extinction coefficients spectrograms of ZnSe at 373 and 423K. From the figure, it is clearly stated that most extinction coefficients of ZnSe at 373 K are close to these values at 423K. It is also seen in the Fig.6 and Fig.3 that the extinction coefficients of ZnSe decrease with the temperature increasing, for example at the wavelength 3 μ m, the extinction coefficient of ZnSe is 3.44E-06, 3.36E-06 and 2.95E-06, at room temperature, 373 and 423K, respectively. As shown in Fig.7, most refractive index values of ZnSe at 373 and 423K fluctuate between 2.2 and 2.4 in the measured spectral range, and the refractive index of ZnSe increases with the temperature increasing, which indicates that the reflectance of ZnSe slab increases with the temperature increasing, because the transmittance and absorptance of ZnSe slab both decrease with the temperature increasing. The reason is that the reflectance of ZnSe slab is a direct proportion function of the refractive index and extinction

coefficient, which indicates that the refractive index of ZnSe is a key parameter to determine optical transmittance performance of ZnSe slab.

For illuminating the deviation between the experimental and the simulated transmittance calculated with deducted optical constants, a parameter of relative error is defined as

$$er(\lambda) = \left| \frac{T_{\text{cal}}(\lambda) - T_{\text{exp}}(\lambda)}{T_{\text{exp}}(\lambda)} \right| \times 100\% \quad (9)$$

Where, $er(\lambda)$ is the relative error. $T_{\text{cal}}(\lambda)$ and $T_{\text{exp}}(\lambda)$ are the simulated and the experimental transmittance values in wavelength λ , respectively.

Fig. 8 shows the relative error between the experimental and the simulated transmittance. It is clearly found that the spectrums of the relative error in the most wavelength region are flat except for some limited wavelength points. As depicted in Fig. 8, most relative errors are lower than 0.1% in the wavelength 2-15 μm , however the relative errors near to the wavelength 4.22 and 15 μm are beyond 0.5%, in which the highest one is 1.47%.

3.3 Optical Properties of ZnSe Slab

The optical constants obtained in section 3.2 are adopted to the optical properties of single and double ZnSe slabs in the wavelength 2-15 μm including reflectance, transmittance and absorptance, which are shown in Fig.9, Fig.10 and Fig. 11.

Fig. 9 shows the transmittance spectrum of single and double ZnSe slabs, in which the thickness of ZnSe slabs is 1, 2, 3, 4, 5 and 6 mm, respectively. From Fig. 9, the shapes of transmittance spectrum of single and double slabs are similar, and the

transmittances of single and double slabs are both decreasing with the slab thickness and temperature increasing. It is also clearly seen that the transmittance performance of single ZnSe slab is much higher than double slabs, and the transmittance difference between single and double ZnSe slabs is more violent with the slab thickness increasing, for example the transmittance ratio between single and double slabs, with thickness 1, 2, 3, 4, 5 and 6mm, is 1.29, 1.32, 1.34, 1.37, 1.39 and 1.42 at 373K and 2.02 μ m, respectively.

Fig. 10 displays the reflectance spectrum of single and double ZnSe slabs, in which the thickness of ZnSe slabs is 1, 2, 3, 4, 5 and 6 mm, respectively. As shown in Fig.10, the shapes of reflectance spectrum of single and double ZnSe slabs are also similar, and the reflectance values of single and double ZnSe slabs are both decreasing with the slab thickness and temperature increasing. It is also clearly seen that the reflectance of double ZnSe slabs is much higher than single ZnSe slab, and the difference of reflectance performance between single and double ZnSe slabs is also more violent with the slab thickness increasing.

Fig. 11 plots the absorptance spectrum of single and double ZnSe slabs, in which the thickness of ZnSe slabs is 1, 2, 3, 4, 5 and 6 mm, respectively. As shown in Fig.11, the shapes of absorptance spectrum of single and double ZnSe slabs are similar, and the absorptance values of single and double ZnSe slabs are both increasing with the slab thickness increasing, but decreasing with the temperature increasing. It is also clearly seen that the absorptance of double ZnSe slabs is much higher than ZnSe slab, and the difference of absorptance performance between single and double ZnSe slabs

is more violent with the slab thickness increasing.

4. Conclusions

Spectroscopic method for determining the optical properties of ZnSe slab was developed and validated. The transmittance spectrograms of ZnSe slabs in the wavelength 2-15 μm were measured. The shapes of transmittance spectrum of ZnSe slabs at 373 and 423K are similar, and these ZnSe slabs all have a wide transparent region.

In the measured spectrums, with the temperature increasing, the extinction coefficient of ZnSe decrease, but the refractive index of ZnSe increases. The spectrum profiles of single and double ZnSe slabs are very similar in shape, and the shapes of the spectrum are not relevantly affected by the slab thickness. However, in the same wavelength, the thicker the slabs layer, the weaker the transmittance and reflectance performance of single and double ZnSe slabs, and the stronger the absorptance performance of single and double ZnSe slabs.

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Figure captions

Fig. 1. Experimental system [28]

Fig. 2. Transmittances of single ZnSe slab at room temperature

Fig. 3. Measured and published values of extinction coefficients of ZnSe

Fig. 4. Measured and published values of refractive index of ZnSe

Fig. 5. Transmittances of ZnSe slabs at 373 and 423K

Fig. 6. Extinction coefficients of ZnSe at 373 and 423K

Fig. 7. Refractive indexes of ZnSe at 373 and 423K

Fig. 8. Relative errors between experimental and simulated transmittances

Fig. 9. Transmittance spectrum of single and double ZnSe slabs(Single: a-373K and
b-423K; Double: c-373K and d-423K)

Fig. 10. Reflectance of single and double ZnSe slabs(Single: a-373K and b-423K;
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Fig. 11. Absorptance of single and double ZnSe slabs(Single: a-373K and b-423K;
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