OPTICAL PROPERTIES OF THERMALLY DEPOSITED BISMUTH TELLURIDE IN THE WAVELENGTH RANGE OF 2.5–10 μm

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The optical constants (the refractive index n, the absorption index k and the absorption coefficient α) of Bi₂Te₃ thin films were determined in the wavelenght range of 2.5 to 10 μm. The shape of the absorption edge in Bi₂Te₃ thin films has been determined from transmittance and reflectance measurements. The edge is of the form expected for direct transition corresponding to $E_g = 0.21$ eV. The optical constants were used to determine the high frequency dielectric constant $\varepsilon_0 = 58$, the optical conductivity $\sigma = \sigma_1 + \sigma_2$ as well as the volume and surface energy loss functions. All these parameters were used to get some information about the intraband and interband transitions.

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1. Introduction

Bismuth telluride has received considerable attention, because of its high thermoelectric properties. These studies were mainly carried out on bulk samples [1], but no much work has been reported on thin films of Bi₂Te₃. So far very few measurements of the optical properties have been reported [2, 3]. Black deduced an optical energy gap of 0.15 eV, while Gibson reported another value of $E_{\rm g}=0.3$ eV.

In the present work we are going to determine the optical constants (n, k and α). The shape of the absorption edge have been studied, since this can lead to useful information concerning the energy band structure as well as the lattice static dielectric constants, and the allowed direct interband transitions.

2. Experimental procedure

Bismuth telluride used in the present work was supplied by Balzers company (West Germany) of purity 99.999%. Bi₂Te₃ films of different thicknesses (25–99.5 nm) were deposited in vacuum of 10⁻⁵ torr, onto potassium bromide single crystals as substrates, using thermal evaporation technique. The rate of deposition was kept constant at 3 nm/s. All the films used in this study were prepared at approximately constant substrate temperature (not exceeding 50°C). To

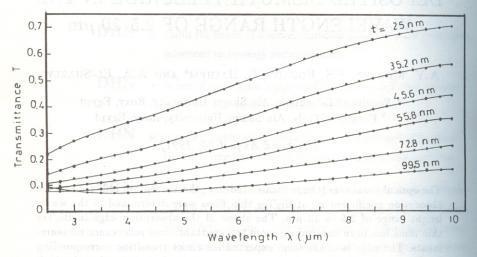


Fig. 1. The spectral dependence of $T(\lambda)$ for Bi₂Te₃ thin films of different thicknesses in the wavelength range of 2.5-10 μ m.

ensure uniform thickness the substrate holder was rotated during the deposition process. The film thickness was measured by applying Tolansky's method [4] using multiple beam Fizeau fringes. The transmittance and reflectance of the prepared samples were measured at normal incidence in the spectral range 2.5–10 μ m using Pyeunican 3P3–300 IR spectrophotometer. Figures 1 and 2 illustrate the transmittance and reflectance in the wavelength range 2.5–10 μ m for Bi₂Te₃ samples of thicknesses between 25 and 99.5 nm.

3. Results

3.1. The optical constants of Bi₂ Te₃ thin films

In order to obtain the optical constants n and k the computational program suggested in [5, 6] was used to solve Murmann's exact formulas [7], representing both transmittance T and reflectance R measured at normal incidence. Figure 3 shows the spectral distribution of both n and k for Bi_2Te_3 thin films of different thicknesses in the spectral range of 2.5-10 μm . The vertical bars in this figures

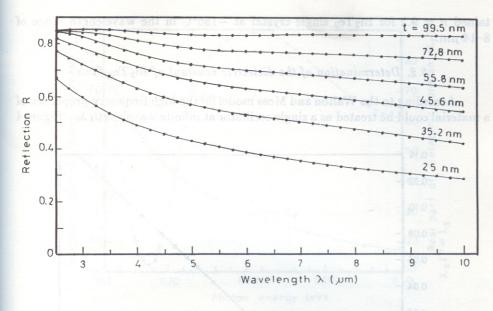


Fig. 2. The spectral dependence of $R(\lambda)$ for Bi₂Te₃ thin films of different thicknesses in the wavelength range of 2.5-10 μ m.

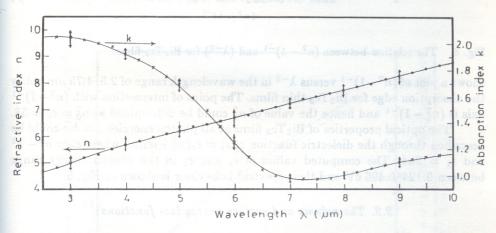


Fig. 3. The spectral distribution of both n and k for Bi₂Te₃ thin films of different thicknesses in the spectral range of 2.5–10 μ m.

represent the discrepancy in the calculated values of n and k of a set of films in the thickness range of 25-99.5 nm. One can see that this discrepancy is relatively small in comparison with the mean value in the whole range of wavelengths. The obtained results indicate that both parameters were independent on the film thickness. It is clear from the figure that the value of n increases from 4.7 to 8.8 through the whole wavelength range used in our investigation. Austin [8] has ob-

tained n=9.2 for Bi₂Te₃ single crystal at -155° C in the wavelength range of $8-14~\mu m$.

3.2. Determination of the dielectric constant of Bi2 Te3 films

According to the Walton and Moss model [9] the high frequency properties of a material could be treated as a single oscillator at infinite wavelength λ_0 . Figure 4

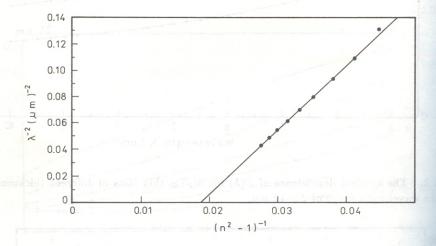


Fig. 4. The relation between $(n^2 - 1)^{-1}$ and (λ^{-2}) for Bi₂Te₃ films.

shows a plot of $(n^2-1)^{-1}$ versus λ^{-2} in the wavelength range of 2.5-4.75 μ m below the absorption edge for Bi₂Te₃ thin films. The point of intersection with $(n^2-1)^{-1}$ axis is $(n_0^2-1)^{-1}$ and hence the value of n_0^2 could be determined as $n_0^2 = \varepsilon_0 = 58$.

The optical properties of Bi_2Te_3 films at all photon energies $\varepsilon = h\omega$ could be described through the dielectric function $\varepsilon(\omega) = \varepsilon_r(\omega) + \mathrm{i}\varepsilon_I(\omega)$ where $\varepsilon_r = n^2 - k^2$ and $\varepsilon_I = 2nk$. The computed values of ε_r and ε_I in the photon energy range between 0.124–0.496 eV and their spectral behaviour is shown in Fig. 5.

3.3. The volume and surface energy loss functions

The volume energy loss function and the surface energy loss function are related to the real and imaginary parts of the dielectric constants by the relations [10]:

$$A = \frac{\varepsilon_{\rm I}}{\varepsilon_{\rm r}^2 + \varepsilon_{\rm I}^2},$$
(1)

$$B = \frac{\varepsilon_{\rm I}}{(\varepsilon_{\rm r} + 1)^2 + \varepsilon_{\rm I}^2}.$$
 (2)

The spectral behaviour of the volume and surface energy loss functions is shown in Fig. 6.

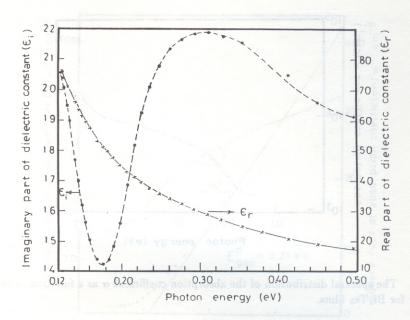


Fig. 5. The spectral behaviour of $\varepsilon_{\rm I}$ and $\varepsilon_{\rm r}$ of Bi₂Te₃ films.

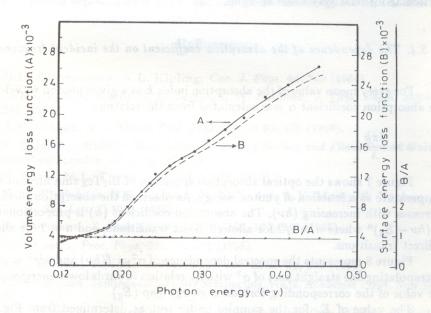


Fig. 6. The spectral behaviour of the volume and the surface energy loss functions of Bi₂Te₃ films.

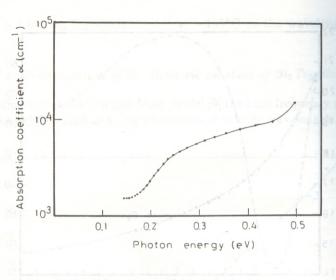


Fig. 7. The spectral distribution of the absorption coefficient α as a function of photon energy for Bi₂Te₃ films.

It is clear that the energy loss by the free charge carriers when traversing the bulk material has approximately the same value when they traverse the surface in particular for relatively lower energies.

3.4. The dependence of the absorption coefficient on the incident frequency

For every mean value of the absorption index k at a given photon wavelength the absorption coefficient α was calculated from the relation:

$$\alpha = \frac{4\pi k}{\lambda}.\tag{3}$$

Figure 7 shows the optical absorption spectrum of Bi₂Te₃ thin films at room temperature as a function of photon energy. As observed the absorption coefficient increases with increasing $(h\nu)$. The absorption coefficient (α) is proportional [11] to $(h\nu - E_g)^n$ where n = 1/2 for allowed direct transitions, and n = 2 for allowed indirect transitions.

Figure 8 represents the spectral dependence of $\alpha^2 = f(h\nu)$ and $\alpha^{1/2} = g(h\nu)$. Extrapolating the straight part of α^2 with $h\nu$ relation towards lower energies yields the value of the corresponding forbidden energy gap (E_g) .

The value of $E_{\rm g}$ for the samples under test as determined from Fig. 8 is 0.21 eV which is in good agreement with the following published data: 0.13 [8], 0.15 [2], 0.30 [3] and 0.16-0.3 eV [12]. The tail in $\alpha^2 = f(h\nu)$ may be attributed to phonon interaction.

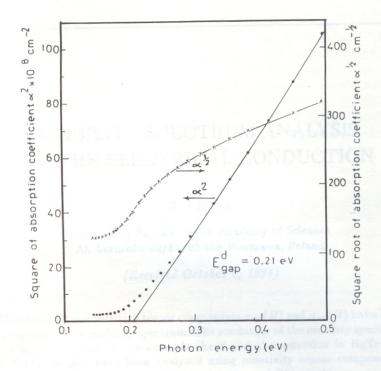


Fig. 8. Spectral dependences of $(\alpha)^{1/2}$ and $(\alpha)^2$ on photon energy for Bi₂Te₃ films.

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