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Differences between surface and bulk refractive indices of a-In_xSe_{1-x}

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Abstract

Thin films of amorphous indium selenide compounds (a-In_xSe_{1-x}) are important, e.g. for photovoltaics. The feature of merit in such applications is also the real part of refractive index n of this material. The data on n in literature are divergent. In this paper, the results of investigations on n in the bulk as well as in the interface layers of thin films of a-In_xSe_{1-x} are presented. The measurements had been performed using optical transmittance and reflectance in spectral range from 1.24 to 1.96 eV of linear polarized radiation that hit the samples with angles of incidence from 0° to 80°. Investigations had been done for sample temperatures from 80 to 340 K. It was found that the refractive index for areas at the free surface n_f is bigger than the refractive index n_b at the interface of thin film–substrate. The averaged over thin film thickness value of real part refractive index $\bar{n}$ have the biggest value in all spectral range. Values of these coefficients increase with increasing the temperature.
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1. Introduction

Thin films of the indium–selenide system (In_xSe_{1-x}) are prospective in the field of optoelectronics, e.g. for constructing solar cells [1,2] and switching devices [3]. They are semiconducting with the energy gap dependent on the molar compositions x in the range from E_g(Se) = 1.85 eV for $x = 0$ [4] to $E_g(In_xSe_{1-x})$ = 0.8 eV for $x = 0.59$ [5]. Another one of the figures of merit of a semiconductor applied in optoelectronics is the value of the real part of its refractive index. Unfortunately the literature data on this important parameter are very different (see, e.g. [6–8]). Some explanation of these differences can be given taking into account the possible inhomogeneities of the investigated films. It is known that the chemical composition of the bulk of a semiconducting compound thin film can be different from the composition near it surface. This difference is affected by the technological processes during the deposition as well as by the adhesion and absorption of gases at the real free surface of the film.

The aim of this paper was to investigate the spectral and temperature characteristics of the real part of refractive index in the bulk as well as near the free surface and near the thin film–substrate interface of In_xSe_{1-x} thin films with different composition. The state of the surface can be taken into account as a result of some surface passivation.

2. Experiment

Thin films of In_xSe_{1-x} had been evaporated on BK-7 substrates in vacuum (10^{-3} Pa). The evaporation had been using previously prepared sources of In_xSe_{1-x} with different molar compositions. The substrate temperature during evaporation was 300 K. Details of the experimental set-up and the technology are given in [9]. Thickness of the obtained films was measured using the standard interference microscopy technique. Chemical composition of the investigated films was determined by X-ray microanalysis. The electron scanning microscope JSM35 was used together with the spectrometer of X radiation with the dispersion of energy LINK860. The molar composition x of the investigated films ranged from 0.25 to 0.69. The structure of the films was confirmed as diffractionally amorphous using the standard X-ray diffraction method.

The optical transmittance and reflectance were measured using PC2000 Ocean Optics Inc. spectrophotometer with master and slave cards with 600 lines grating (blazed at 500 and 400 nm, respectively). The spectrophotometer was equipped

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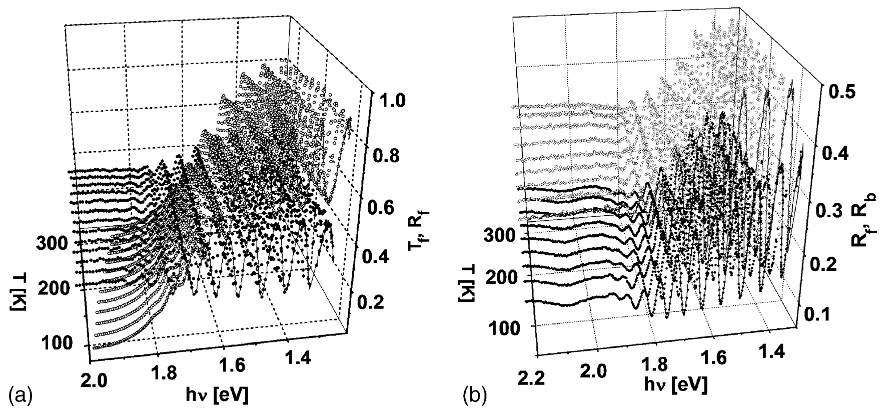

Fig. 1. (a) Optical transmittance T_f (○) and reflectance R_f (●) of $In_{0.55}Se_{0.45}$ $w = 1.89\,\mu m$ thick film illuminated perpendicularly from the free surface side; (b) comparison of the optical reflectances of $In_{0.55}Se_{0.45}$ thin film illuminated perpendicularly from the free surface side R_f (○) and from the substrate side R_b (●).

with the DH2000-FHS deuterium–halogen light source from Ocean Optics Inc. For the normal incidence of light the spectrometer was equipped with Y-type waveguide (R 400x7–2-HOH-M—Ocean Optics) for reflectance measurements and single waveguide (SMA-600-2HOH—Ocean Optics) for transmittance measurements. For the non-normal incidence of light two waveguides (P 600-2-UV/VIS—Ocean Optics) were used with goniometer (GUR-5 LOMBO). One of the waveguides was equipped with collimating lens (84-UV-25—Ocean Optics). The Glan-Thomson prism was between the collimated output from the source waveguide and the sample. The sample was mounted in 1.33 Pa vacuum in the optical D2209 chamber of R2205 Cryogenic Microminiature Refrigeration II-B System was based on Joule Thomson effect (MMR Technologies, Inc.). This system was controlled with certainty 0.1 K using K7701 temperature controller. It allowed temperature selection over the range 80–333 K without any need for liquid nitrogen participation. High-pressure (12 MPa) nitrogen gas was expanded through small capillaries in the device producing liquefied nitrogen under the cold pad. The temperature of the pad was controlled by a resistive heater and a silicon-diode temperature sensor built into the pad. The multiple averaged spectral characteristics containing 2048 data points for different wavelengths were registered in various temperatures using the OOI-Base program from Ocean Optics Inc. The measurements were performed for different polarization of the radiation incident upon the samples as well as for different angles of incidence.

Fig. 1 presents some typical for the investigated samples temperature and spectral characteristics measured for the $In_{0.55}Se_{0.45}$ $w = 1.89\,\mu m$ thick film. Fine interference fringes are evident in the spectral characteristics of optical transmittance and reflectance for smaller photon energies, however their positions depend on the temperature due to the temperature dependence of the real part of refractive index of the investigated material. From the evident decrease of optical transmittance one can evaluate the absorption edge and consequently the value and temperature dependence of the optical energy gap of the material. The data recorded in the cases of incidence of light on the free surface and on the back surface of the sample are different, especially for optical reflectance. Fig. 2 presents typical changes in spectral characteristics due to the various angles of incidence of plane polarized radiation upon the sample. The angular dependences of the transmittance and reflectance for plane polarized radiation with electric vector parallel and perpendicular to the plane of incidence are different. Although in every case a similar influence of the angle of light incidence on the position

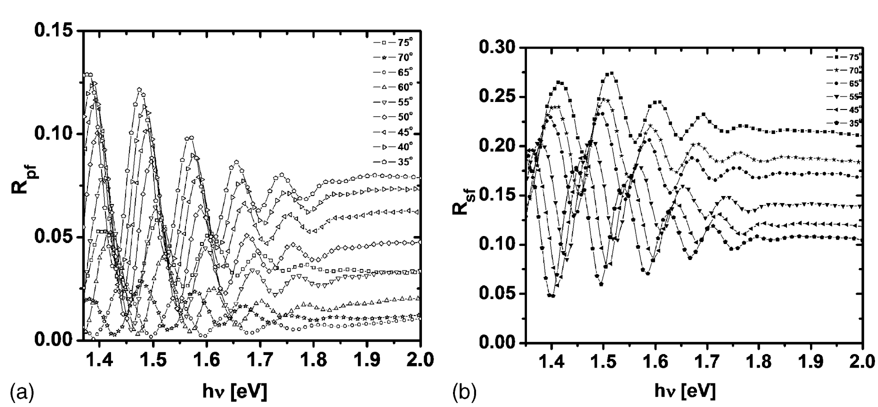

Fig. 2. Spectral characteristics of optical reflectances of $In_{0.41}Se_{0.59}$ $w = 1.24\,\mu m$ thick film for different angles of incidence of plane polarized light up to the free surface (a: plane polarized radiation with electric vector parallel to the plane of incidence R_{pf}; b: plane polarized radiation with electric vector normal to the plane of incidence R_{sf}; $T = 293$ K).

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