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Physica B 366 (2005) 38–43
PHYSICA B
www.elsevier.com/locate/physb

Electrical conductivity and crystallization kinetics of amorphous $Se_{0.81}In_{0.19}$ films

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Received 18 May 2004; received in revised form 4 March 2005; accepted 10 May 2005

Abstract

The temperature dependence of the DC conductivity (σ) of amorphous Se and $Se_{0.81}In_{0.19}$ thin films, prepared by thermal evaporation, has been studied. The incorporation of In atoms in a Se matrix leads to an increase in the electrical conductivity of $Se_{0.81}In_{0.19}$ and to a decrease in the thermal activation energy of conduction from 0.57 to 0.24 eV.

The change in σ with time during the amorphous-to-crystalline transformation is recorded for $Se_{0.81}In_{0.19}$ films at different isothermal temperatures in the range 343–373 K. The results indicate that the micro-heterogeneous structures of the films have a remarkable influence on the electrical conductivity during the amorphous-to-crystalline transition. The formal crystallization theory of Avrami has been used to calculate the kinetic parameters of crystallization. The activation energy of the amorphous-to-crystalline transformation is found to be 0.82 and 1.29 eV, respectively, for the two stages of crystallization during the time period of the transformation process.

PACS: 61.34.Fs; 64.70.Pf; 72.20.Ee; 72.80.Ng

Keywords: Amorphous semiconductors; Chalcogenides; Electrical properties; Crystallization kinetics; Selenium–Indium

1. Introduction

The layered semiconductors are perspective materials because of their potential applications arising from their particular structure. Among these, the Se–In ones have valuable optical and electrical properties, making their thin film forms attractive to be used as low-cost materials in solar cell applications (Refs. [1,2] and references therein). In a previous paper [3], we showed that an ice-water-quenched ingot of $Se_{0.9}In_{0.1}$ has a stable phase consisting of some crystalline clusters embedded in a glassy matrix.

It is well known that if a non-crystalline material is annealed at a temperature below its

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doi:10.1016/j.physb.2005.05.018

glass transition temperature (T_g), the structure approaches a metastable equilibrium and the corresponding macroscopic properties such as enthalpy or volume, etc. change with time. The phase transition of non-crystalline materials occurs when an undercooled liquid is annealed well above T_g, which is called crystallization, on reheating. There are numerous papers published on the glass transition, structural relaxation and crystallization phenomena in non-crystalline materials and their number has increased steadily during the last three decades (Refs. [4–14] and references therein). Although different structural-sensitive physical parameters as well as advanced thermal analyses are frequently used under isothermal and/or non-isothermal conditions, it is still not so easy to explore the mechanism of these rather complicated processes and related properties of non-crystalline materials. However, all these studies are mostly carried out on bulk glassy materials and very rarely, to the best of our knowledge, on amorphous thin film forms.

The aim of this paper is to demonstrate the possibilities of crystallization kinetic studies in amorphous thin films by the time dependence of electrical conductivity as illustrated by using binary amorphous semiconductor thin Se–In films prepared by thermal evaporation from $Se_{0.9}In_{0.1}$ bulk form.

2. Experimental details

Bulk material of the composition $Se_{0.9}In_{0.1}$, previously prepared by a conventional quenching technique [3], was used as a ground material for preparing the Se–In films investigated. The films were prepared by the thermal evaporation technique using vacuum coating unit type Edwards E306A. Pre-cleaned corning and quartz glass substrates were fixed onto a horizontal rotating holder (with rotation speed of 4 cycles/min) to obtain films homogeneous in thickness and composition. All the films used were deposited in the same evaporation run. The distance separating the evaporated source material and the substrate was ~14 cm. The temperature of the substrates was kept constant at room temperature (RT). The base pressure of the vacuum system was 4×10^{-6} Torr. During evaporation, the vacuum pressure was stable at 1.5×10^{-6} Torr and the deposition rate was in the range of 1–2 nm/s. The film thickness was monitored using a thickness monitor model Edwards FTM5. After evaporation, the thickness of the fresh films was accurately determined by an optical interference method.

Identification of the structural phase as well as the chemical composition of the deposited films was determined using X-ray diffraction (XRD) with a CuK_α radiation source, and a scanning electron microscope (SEM) using a computerized Philips EXPERT-MPDUG PW-3040 diffractometer and a Philips XL-130 SEM attached to an energy-dispersive X-ray analyzer (EDAX), respectively. The EDAX analysis was performed at four different locations on the surface of the prepared films, thus ensuring the composition homogeneity of each investigated film. The XRD pattern of the deposited films is characterized by the absence of any diffraction peaks indicating the amorphous nature of the prepared films. The actual thin film composition of the deposited films, as identified by EDAX, is found to be $Se_{0.81}In_{0.19}$.

A convenient two-probe method was used to measure the direct current (DC) conductivity of the investigated films. Electrical contacts were achieved by the thermal evaporation of gold electrodes in a coplanar configuration of separation ~2 mm. The current–voltage measurements showed that the applied contacts were ohmic. The temperature dependence of conductivity was handled in a steady-state condition in the temperature range from $T < T_g$ (320 K, [3]) down to liquid N_2. The time dependence of conductivity was measured under isothermal condition at different temperatures around T_c (the onset of the exothermic crystallization peak on the DTA curve for bulk $Se_{0.9}In_{0.1}$; 361 K at 10°/min, [3]), and below the endothermic melting point T_m (473 K, [3]). All conductivity measurements were done under vacuum using an Oxford cryostat (type Oxford DN-1714) connected to an Oxford ITC-503 temperature controller of accuracy 0.01°, and a Keithley-616 digital electrometer.