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Effect of doping by transitional elements on properties of chalcogenide glasses

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Abstract

In present work, the results on the influence of doping by transitional elements on thermal, optical, structural and magnetic properties of chalcogenide glasses are presented.

Thermal properties (T_g values for undoped and doped glasses) were studied using differential scanning calorimetry technique. Activation energy of glass transition was estimated with the use of Kissinger's expression. Structural studies were carried with the use of Raman and infrared spectroscopy and X-ray diffraction. Radial electron distribution functions in doped and undoped bulk glasses were obtained and analyzed. In Raman spectra, main observed effect under the introduction of dopants was the change of relative concentration of main and non-stoichiometric structural units characteristic for As_2S_3 glasses. Investigation of influence of transition metals Mn-dopants on the optical properties of As_2S_3 glass was studied in mid-IR region. Pure chalcogenide glasses are diamagnetics. Introduction of transitional and rare earth impurities changes the magnetic properties of investigated chalcogenide glasses.
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1. Introduction

Chalcogenide glasses (ChGs) exhibit a number of interesting optical properties with various potential applications as reviewed in, for example [1–5], etc. As frequently pointed out by various researchers, ChGs are promising materials for various applications because they are transparent over a wide range of wavelengths in the infrared (IR) region, they possess high refractive indices, low phonon energies and are easy to fabricate. Chalcogenide glasses can be used in applications in sensorics, infrared optics and optoelectronics. The glasses can be used for the preparation of optical fibers both for passive and active applications. Developments in photonics applications have highlighted the chalcogenide glass as a host for rare-earth elements.

The refractive index and its wavelength dependence, other optical properties are among important parameters that determine the suitability of materials as optical media. Refractive and absorption indexes, optical band gap of chalcogenide glasses can be changed by doping of different elements [3–6].

Special interest for applications is related with chalcogenide glasses doped with optically active rare-earth and transition metal ions, because they alter electrical, thermophysical, mechanical, magnetic and optical properties of the host material due to structural and electronic changes of the glass network [6–8].

The present work was devoted to studies of the influence of doping by transitional elements (Mn) on thermal, optical, structural and magnetic properties of As_2S_3 chalcogenide glasses.

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2. Experimental

The As_2S_3 glasses with manganese concentration from 0 up to 5 wt% Mn were prepared by standard melt-quenching technique using constituent elements of 6 N purity in vacuum-sealed silica ampoules. Ampoules were heated at 80 K/h rate, melt was hold at 1010 K during 80 h with subsequent quenching in the air at 10 K/h rate. Glass samples were obtained in the form of bar with 10 mm diameter. The prepared bulk glasses were cut into plates of 1 mm in thickness and polished to yield samples with high quality flat surfaces suitable for optical measurements.

X-ray diffraction measurements (XRD) were carried out using ARL X'tra (Thermo scientific) installation. Cu K_λ radiation (λ=0.154 nm) was used. Diffraction spectra were recorded by θ–θ-scanning in 2–140° range at room temperature. Measurements were carried out in step regime with scanning step 0.2° and point acquisition time 5 s.

Room temperature Raman spectra were recorded using Fourier spectrophotometer Bruker IFS-55 Equinox with FRA-106 attachment (with measurement step 1 cm^1). Nd:YAG laser light at 1.06 μm wavelength was used for excitation.

The positron annihilation lifetime spectrum was taken by conventional fast–fast coincidence method using plastic scintillators coupled to Phillips XP2020 photomultipliers with a ^{22}Na source placed between two sandwiched samples. The time resolution (FWHM) was about 320 ps, measured by defect free Al sample with 3 Gaussian approximation of resolution function. A total number of coincidences in analyzed time spectrum overreach 1×10^6 counts. Analysis of lifetime spectrum was carried out using the LT-9.0 program of Kansy [9].

Room temperature transmission spectra in the 700–4000 cm^{-1} region were recorded using an FT spectrometer "Perkin Elmer" Spectrum BXII.

Thermal properties were studied using differential scanning calorimetry (DSC) technique, T_g values for undoped and doped glasses were obtained. NETZSCH DSC 404 (with accuracy ± 0.5 K) calorimeter was used in DSC measurements. Calorimetric measurements were carried out using powder samples ($m \sim 20$ mG) in argon atmosphere under temperature changes within 40–250 °C. Heating rate consisted $q = 10$ K/min. Calibration of calorimeter was carried out by melting of pure metals In, Sn, Bi, Pb, Al, Cu with known values of temperature and enthalpy of melting.

Magnetizations of samples were measured with Cryogenic S600 Super-conducting Quantum Interference Device (SQUID) magnetometer in the temperature range of 5–400 K and in magnetic fields up to 6 T. A cryogenic system is completed with automated instrument control, data acquisition and analysis using the National Instrument's LabVIEW software.

3. Results and discussion

3.1. XRD measurements

X-ray diffraction patterns (Fig. 1) confirm the amorphous nature of the bulk samples of chalcogenide glasses. Radial electron distribution functions (Fig. 2) in doped and undoped bulk glasses were obtained and analyzed [10].

3.2. Raman spectra

From Fourier Raman spectra information (Fig. 3) on the structural changes in As–S glasses doped with transitional metals was obtained. Introduction of such dopants leads to the concentration increase of non-stoichiometric molecular fragments (such as As_4S_4, etc.).

Introduction of manganese leads to the intensity increase of 192, 227, 236, 365 cm^{-1} bands that correspond to the vibrations of non-stoichiometric molecular fragments As_4S_4. Intensity of band near 496 cm^{-1}, characteristic for vibrations of S–S bonds is decreasing. In 130–190 cm^{-1} range bands appear, that can be connected with the creation of new sulphur containing structural units, similar to MnS molecular fragments [6,11]. The main feature of Raman spectra under the introduction of manganese

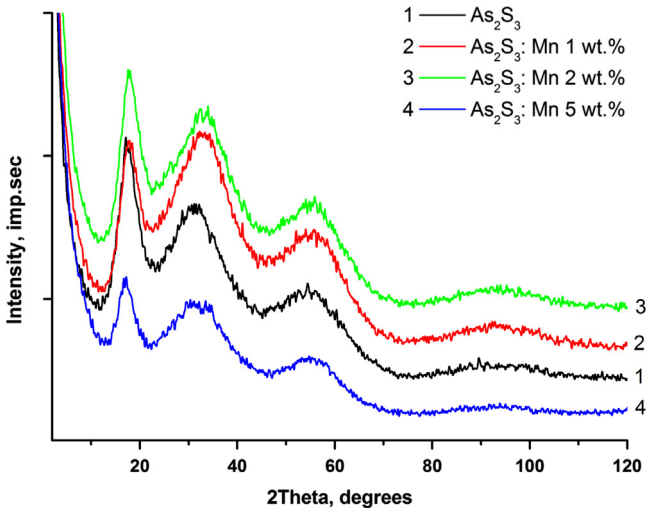

Fig. 1. X-ray diffraction patterns of As_2S_3 doped by Mn. Spectra are shifted on equal distance in order of appearance.

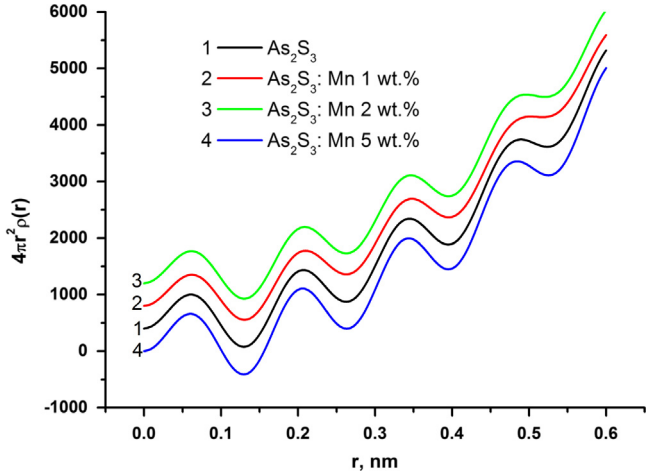

Fig. 2. Curves of radial electron distribution function of As_2S_3 doped by Mn. Curves are shifted on equal distance in order of appearance.

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