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Optical tomography of non-crystalline films
by interference enhanced Raman spectroscopy

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Abstract The depth dependence of Raman spectra of a-GeS₂-type films having a different optical thickness ($\lambda/4$ and $\lambda/2$) and their refractive index profile have been investigated. The model of a layered-inhomogeneous structure of films has been proposed. There have been distinguished three regions: near-surface region (up to 50 Å), central part and transition film-substrate region (up to 300 Å).

Introduction

To overcome obstacles connected with the insufficient intensity of the Raman signal for structural investigations of thin and superthin films, the principles of adaptive optics have been used [1, 2]. Their use for interference enhancement of Raman signal (IERS) allows one to redistribute the field intensity of an exciting light wave (E^2) inside a three-layered interference structure. The possibility to redistribute E^2 in the film is especially valuable while studying depth dependences of physical properties of thin films by optical methods within optical tomography [2]. The aim of the present work is to develop an IERS method to investigate depth dependences of structural peculiarities for thermally evaporated thin films of strongly dissociative a-GeS₂-type glasses.

Experimental

The a-GeS₂-SiO₂-Al three-layered structure was prepared by flash evaporation of GeS₂ powder onto a substrate with preliminarily deposited Al and SiO₂ (1600 Å) by conventional methods. The thickness of a-GeS₂-type films during deposition was controlled by

photometric methods. Raman spectra were taken with a DFS-24 instrument with a microoptical attachment in the geometry of back scattering of reflection. As the exciting source for the Raman signal a set of lasers with wavelengths of 4800, 6300 and 7500 Å was used. The distribution of the refractive index profile in films was studied by multiangular ellipsometry.

The distribution of the field in a light wave in the upper high-refractive layer of an a-GeS₂-SiO₂-Al structure was calculated using the equation given in [3].

Results and discussion

The distribution of the field in a-GeS₂ films with an optical depth of $\lambda/4$ and $\lambda/2$ is given in Fig. 1. The analysis of IERS for the near-surface region of As₃(GeS₂)₉₇ films multiple to $\lambda/4$ and $\lambda/2$ [3] ($\lambda_{\text{ex}} = 4800; 7500 \text{ \AA}$) and the data of the present investigations (Fig. 2) show that the position of bands in the region of 200–400 cm⁻¹ is close to the position of bands in Raman spectra of Ge_xSe_{1-x} glasses rich in germanium ($x = 33\text{--}40 \text{ at\%}$, $n = 2.5\text{--}2.6$ at $\lambda = 6300 \text{ \AA}$). The band at 335 cm⁻¹ (Fig. 2) can be related to a position close to the $\nu_1(\text{A})$ band in a-GeS₂, while applying hydrostatic pressure [4], the band at 290 cm⁻¹ coincides with the position of the band in the Raman spectrum of a-Ge [1]. The shift of the $\nu_1(\text{A})$ band in the Raman spectrum of the film into a low-frequency region in comparison with $\nu_1(\text{A}) = 342 \text{ cm}^{-1}$ in a-GeS₂ [3] is supposed to be caused by the availability of internal deformations in the film at the initial growth stage. The availability of near-surface and transition film-substrate regions of structural units (s.u.) rich in germanium is also confirmed by X-ray photoelectronic investigations [3]. The central part of the films similar to bulk glass is formed presumably from GeS_{4/2} s.u. A joint consideration of Raman spectra structural analysis data at different depths of the E^2 maximum penetration into the film at $\lambda_{\text{ex}} = 4800 \text{ \AA}$, and $\lambda_{\text{ex}} = 7500 \text{ \AA}$ [3] (Fig. 2), the solution of back ellipsometric problem and the data of studying SIMS profile [3] allow us to propose the model of a layered-inhomogeneous film structure and its

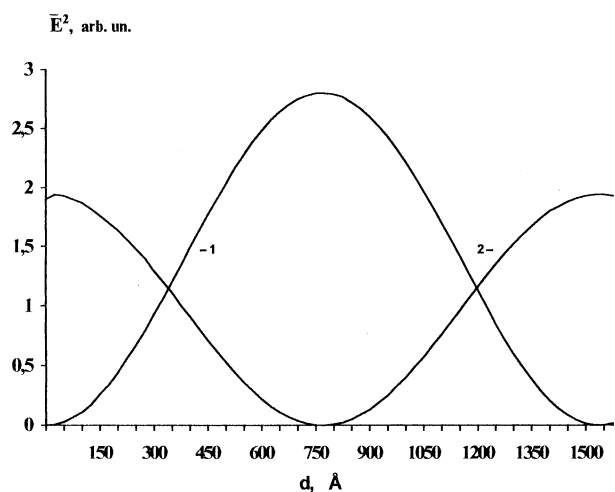


Fig. 1 Depth dependence of the field intensity \bar{E}^2 in $\lambda/2(1)$, $\lambda/4(2)$ films of the a-GeS₂-type on a-GeS₂-SiO₂-Al three-layer structure

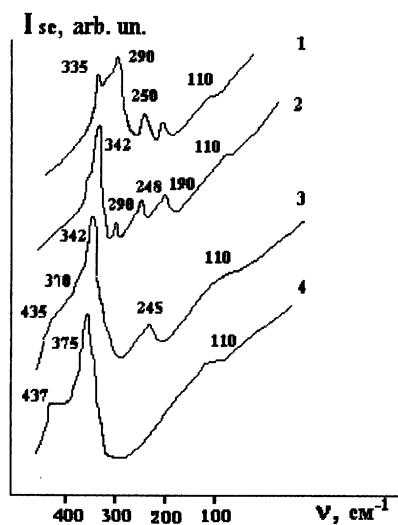


Fig. 2 Depth dependence of IERS Raman spectrum $A_{S_x}(\text{GeS}_2)_{100-x}$ films and glasses $\lambda_{\text{ex}} = 6300 \text{ \AA}$: 1 - $x = 3$, $d = 800 \text{ \AA}$; 2 - $x = 3$, $d = 8000 \text{ \AA}$; 3 - $\text{As}_3(\text{GeS}_2)_{97}$ -glass; 4 - GeS_2 -glass

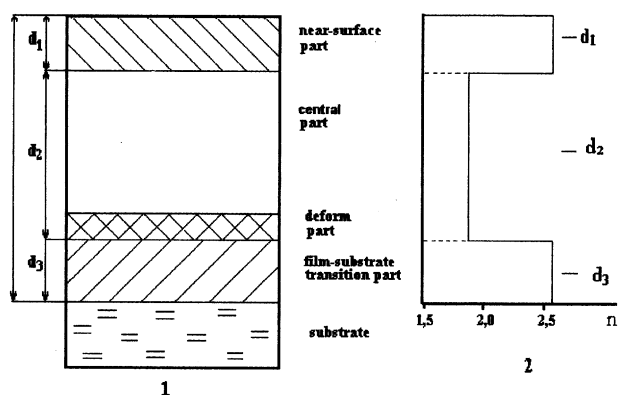


Fig. 3 Model of the non-uniform layer structure for a-GeS₂-type films (1) and model non-uniform profile of refractive index at $\lambda_{\text{ex}} = 6300 \text{ \AA}$ for a-GeS₂ (2)

refractive profile (Fig. 3). The availability of the near-surface layer d_1 (up to 50 \AA) as well as the transition film-substrate layer d_2 with a thickness up to 300 \AA is common to thermally evacuated a-GeS₂-type films.

Conclusion

The structural interpretation of Raman spectra of a-GeS₂-type films at different positions of the \bar{E}^2 maximum in the layer together with the data of SIMS profiles and the depth dependence of refractive indices point to a layered-inhomogeneous distribution of the structure, composition and refractive index profile over the thickness of films prepared by flash evaporation.

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