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Relaxation Processes in Erbium Monosulfide Thin Films

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Abstract.

Thin crystalline ErS films have been grown by flash evaporation in vacuum using a pre-synthesized bulk material on quartz and sapphire substrate. The bulk material was synthesized by direct synthesis in sealed quartz ampoules at a temperature of 800 K. During the film growth process, the vacuum in the deposition chamber was maintained at $\sim 10^{-6}$ Pa, evaporator temperature was ~ 2750 K and the substrate temperature ~ 900 K. Film thickness was about 0.4-0.6 μm. All prepared films had a stoichiometric composition and a NaCl crystal lattice with a lattice parameter a = 5.48 Å. After deposition of films and cooling to room temperature the dependence of electrical resistivity on time was investigated by the four-probe method at atmospheric pressure. It is shown that the relaxation process on films deposited on a quartz substrate proceeds much more slowly than on films prepared on a sapphire substrate. A possible mechanism for such a difference is given.

Keywords: deposition; electrical resistivity; relaxation; thermal expansion; thin film; vacuum-thermal substrate.

Introduction

Rare-earth element (REE) chalcogenides, especially their thin films, are increasingly used in modern technology [1–3], so the study of their physical properties is an urgent task. In addition, of great theoretical interest is the study of the formation of structures of thin films prepared under nonequilibrium conditions [4, 5]. In that work a technology for preparing thin ErS films on various substrates was developed and a study of relaxation processes was carried out by measuring the time

dependence of the electrical resistance. As is known, thin films are convenient objects for studying relaxation processes, since during thermal deposition of a thin film on a substrate, a nonequilibrium film-substrate system is formed, nonequilibrium arises due to the difference in the coefficients of thermal expansion of the film and substrate material, in addition, mechanical stresses arise at the interfaces nanocrystals that form the structure of the films. The combination of these factors causes the possibility of thermodynamic equilibrium processes occurring, which affects the electrophysical parameters of the film, including the electrical resistance.

Main Part

EXPERIMENTAL

Erbium monosulfide films 0.5 μm thick were prepared by flash evaporation in vacuum using a presynthesized bulk material with the composition ErS. The bulk material was synthesized by direct synthesis in sealed quartz ampoules at a temperature of 800 K, During the deposition process, the vacuum in the working chamber was $\sim 10^{-6}$ Pa, evaporator temperature was ~2750 K and the substrate temperature – ~900 K The films were prepared on substrates having the shape of a rectangular parallelepiped with dimensions of 12 × 7 × 0.5 mm made of fused quartz and sapphire. The deposition rate was $\sim 12 \times 10^{-10}\ m/s$. According to X-ray and electron diffraction analysis, single-phase polycrystalline films of the stoichiometric composition ErS with a cubic structure of the NaCl type with a crystal lattice parameter a = 5.48 Å are formed on all used substrates. X-ray microanalysis showed that the films contain 49.8 ± 0.1 at % Er and 50.2 ± 0.1 at % S. According to the Auger spectral analysis, the thickness composition of the films is constant within the experimental error of ± 0.05 at %.

After the films were deposited and cooled in the deposition chamber to room temperature, the electrical resistivity was measured by the four-probe method at atmospheric pressure. The measurement accuracy was 3-4%. During repeated measurements, errors may occur due to the appearance and growth of an oxide film on the surface of the measured film. To minimize this process, the measurements were carried out without detaching the probes from the surface during the entire measurement process. To average the measurement result, 5-7 points on the surface of the films were used. The measurements were carried out on eight films deposited on various substrates.

RESULTS AND DISCUSSION

Fig. 1 shows the dependence of the electrical resistivity on the exposure time of ErS film at room temperature and normal atmospheric pressure, and in Fig. 2 – of ErS film deposited on a sapphire substrate. As can be seen from the figures, with increasing exposure time, the resistivity of the films deposited on both the quartz substrate and on the sapphire substrate decreases in almost the same pattern, the difference is only in the time to reach the saturated state, so the resistance of the film deposited on the quartz substrate decreases from $\sim 5.4 \cdot 10^{-7}\ \Omega \cdot m$ and reaches saturation - ~ $4.8 \cdot \sim 5.4 \cdot 10^{-7}$ Ohm · m within ~148 min, the resistance of the film on a sapphire substrate decreases from $\sim 5.3 \cdot 10^{-7} \Omega \cdot m$ and reaches saturation - $\sim 4.75 \cdot 10^{-7}\ \Omega \cdot m$ within ~110 min. As can be seen from the data presented, the time to reach the saturated state of the films prepared on a quartz substrate is approximately 35% higher than that for films on a sapphire substrate. Additional resistivity measurements were carried out 18 hours after the deposition of the films on both quartz and sapphire substrates. The measurements showed that, within the limits of the measurement error, an increase in the exposure time does not affect the values of the specific saturation resistance, both for films prepared on quartz and sapphire substrates.

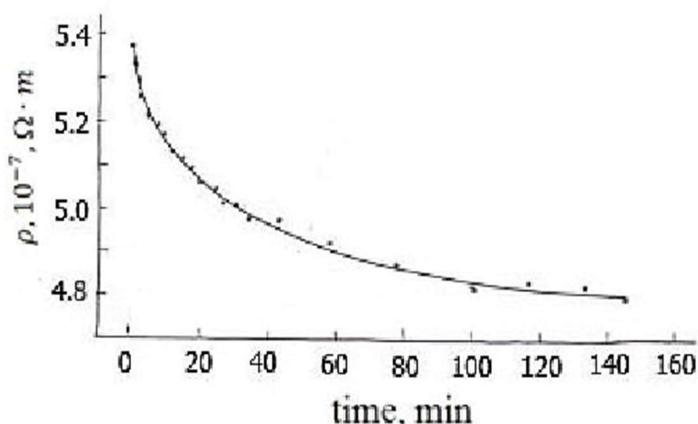

Fig. 1. Dependence of the electrical resistivity of the ErS film on the exposure time
(quartz substrate, film thickness 0.5 μm)

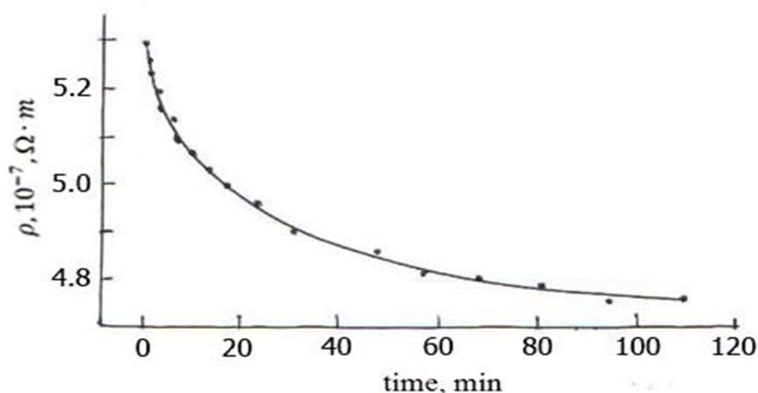

Fig. 2. Dependence of the electrical resistivity of the ErS film on time exposure
(sapphire substrate, film thickness 0.5 μm)

The difference in the time to reach the stationary state of the electrical resistivity of the ErS films, depending on the substrate material, can be associated with differences in the thermal expansion coefficients (TEC) of the ErS–quartz and ErS–sapphire pairs. In the scientific literature, there are no data on the TEC of ErS, given that the TEC of many REE monochalcogenides is in the range 11, 62 $\cdot$ 10^{-6} K^{-1} - 15, 35 $\cdot$ 10^{-6} K^{-1} [6], then we can assume that the TEC of ErS will be of the same order. The TEC of quartz is 0.77-1.4 $\cdot 10^{-6}$ K^{-1} and that of sapphire is 7-10 $\cdot$ 10^{-6} K^{-1} [7]. As can be seen, the TEC of quartz is almost ten times less than the TEC of sapphire; therefore, it can be assumed that the crystal lattice of the ErS film deposited on a quartz substrate will be in equilibrium to a much lesser extent than the film deposited on a sapphire substrate, which is the reason for the increase in the exposure time to reach an equilibrium state.

Conclusion

Thin crystalline ErS films have been grown on quarts and sapphire substrates by flash evaporation in vacuum of presynthesised bulk crystal with composition ErS. At room temperature and atmospheric

pressure, the dependence of the electrical resistivity on the exposure time of films prepared on quartz and sapphire substrates was studied. It is shown that the relaxation time of the electrical resistance of films prepared on a quartz substrate is almost ~35% longer than that of films on a sapphire substrate. It has been suggested that such a difference is associated with the difference in temperatures of thermal expansion of pairs of materials ErS – quartz and ErS – sapphire. In the first case, the difference is much larger than in the second case; accordingly, the mechanical stresses in the film are greater and a longer time is required for relaxation.

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ანოტაცია. წინასწარსინთეზირებული მოცულობითი მასალის დისკრეტული ვაკუუმურ-თერმული აორთქლებით მომზადდა ErS ფირები ნადნობი კვარცისა და ლეიკოსაფირონის ფუძეშრეებზე. ErS მოცულობითი კრისტალები მიღებულ იქნა პირდაპირი სინთეზის მეთოდით კვარცის ამპულებში ~ 800 K ტემპერატურაზე. ErS ფირების ზრდისას კამერაში ვაკუუმი შეადგენდა $\sim 10^{-6}$ პა, ამაორთქლებლის ტემპერატურა იყო ~ 2750 K, ფუძეშრის ტემპერატურა ~ 900 K, დაფენილი ფირების სისქე – 0.4-0.6 მკმ. ყველა ფირს ჰქონდა სტექიომეტრიული შემადგენლობა და NaCl-ის ტიპის მესერი, მესრის მუდმივა იყო 5.48 Å. ფირების დაფენისა და ოთახის ტემპერატურამდე გაცივების შემდეგ, ოთხზონდიანი მეთოდით გაზომილ იქნა კუთრი ელექტროწინაღობის დამოკიდებულება დაყოვნების დროზე ნორმალური ატმოსფერული წნევის პირობებში. ექსპერიმენტებმა აჩვენა, რომ კვარცის ფუძეშრეზე დაფენილ ფირებში რელაქსაციის პროცესი მიმდინარეობს გაცილებით ნელა ლიკოსაფირონის ფუძეშრეზე დაფენილ ფირებთან შედარებით. მოყვანილია ასეთი განსხვავების შესაძლო მექანიზმი.

საკვანძო სიტყვები: ელექტროწინაღობა; ვაკუუმურ-თერმული დაფენა; რელაქსაცია; სითბური გაფართოება; ფირი; ფუძეშრე.

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