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ORIENTATION AND GROWTH OF LEAD SULPHIDE FILMS

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Transmission electron microscopy and diffraction techniques were employed to investigate the growth and orientation of thin films of lead sulphide condensed from the vapour phase onto amorphous and crystalline substrates at 300 K. Films grown on amorphous substrate were polycrystalline. In case of deposition on cleaved NaCl substrate, the deposit shows an oriented growth such that (111) PbS | | (100) NaCl. Effect of electron beam on PbS thin films has been investigated.

1. Introduction

The growth of thin films by deposition from the vapour phase occurs by a nucleation mechanism followed by enlargement and intergrowth of these nuclei to form continuous films. The deposit thickness at which continuity occurs depends markedly on the substrate-deposit combination as well as deposition condition[1] (e. g. deposition rate, substrate temperature). Most nucleation theories explain the role of substrate in determining epitaxial orientation by assuming that the substrate energetically favours the adsorption of one geometrical arrangement of atoms in the cluster or nuclei over another. On the other side, thin films deposited on amorphous substrates are consisted of randomly oriented polycrystals[2].

The single crystal substrate has a dominant influence on the oriented growth of the deposit. However, the resulting orientation of the deposit depends on the orientation and the crystal structure of the substrate[3]. The change in the orientation of thin films with increasing thickness was attributed to surface diffusion and migration of the grain boundaries during coalescence of the two differently oriented nuclei. PbS nuclei appears to form preferentially on surface emergent dislocations or slip steps on the NaCl surface[4]. Matthews[5] has suggested that a high deposition rate can favour epitaxy because of its effect on the coalescence of nuclei. Epitaxial growth is observed for materials of different structures on cubic alkali halides as well as monoclinic mica[6]. The resulting orientation of the deposit depends on the orientation and the crystal structure of the substrate. Some symmetry exists, though not always obvious between the contacting planes of the two materials.

Marlin[7] classified the problem of radiation damage under two headings: the production of lattice defects due to the slowing down of energetic atoms and the eventual disappearance of these defects in relation to thermal migration. The decomposition process[8] consisted of breaking up the single crystalline material into polycrystalline aggregates. At the same time cavities or larger aggregates of vacancies were produced.

2. *Experimental technique*

High purity PbS powder was employed for the deposition of thin films in vacuum (about 10^{-7} Pa) by evaporation from molybdenum boat on amorphous carbon and freshly cleaved surfaces of rock-salt at 300 K. The rate of deposition was estimated to be about 5 nm/s. The films were examined in an electron microscope (ELM 1 D_2) operating at 45 kV. The thickness of the film was determined interferometrically[9]. The thickness of the thin films deposited was changed from 30 to 80 nm.

3. *Results and discussion*

The condensation of vapour atom is determined by its interaction with the impinged surface. The impinging atom is attracted to the surface by the instantaneous dipole and quadrupole moments of the surface atoms. As a result, the atom loses its velocity component normal to the surface in a short time, provided the incident kinetic energy is not too large. The vapour atom is then physically adsorbed called »adatom«, but it may or may not be completely thermally equilibrated. The adatom has a finite stay or residence time on the surface during which it may interact with other adatoms to form a stable cluster and be chemically adsorbed (incorporated into the surface with the release of heat of condensation). If not adsorbed, the adatom reevaporates or desorbs into the vapour phase[10].

A sequence of transmission electron micrographs (Fig. 1) shows the various stages of growth of lead sulphide films deposited on freshly cleaved rock-salt substrate. Rock-salt was as likely as to initiate parallel orientation in PbS because its axial length (0.5639 nm) is within 6% of that of PbS (0.5929 nm). Fig. 1 a shows the film as widely separated nuclei and coalescence of them. Coalescence phenomena have profound effect on the structure and properties of the resultant film since recrystallization, grain growth, orientation and removal of defects etc., occur as a consequence of coalescence[11]. The orientation of PbS films relative to (100) of NaCl is (111). As the thickness increases, the closing stage proceeds and a strong oriented layer is developed as shown in Fig. 1 b, the oriented deposit must arise

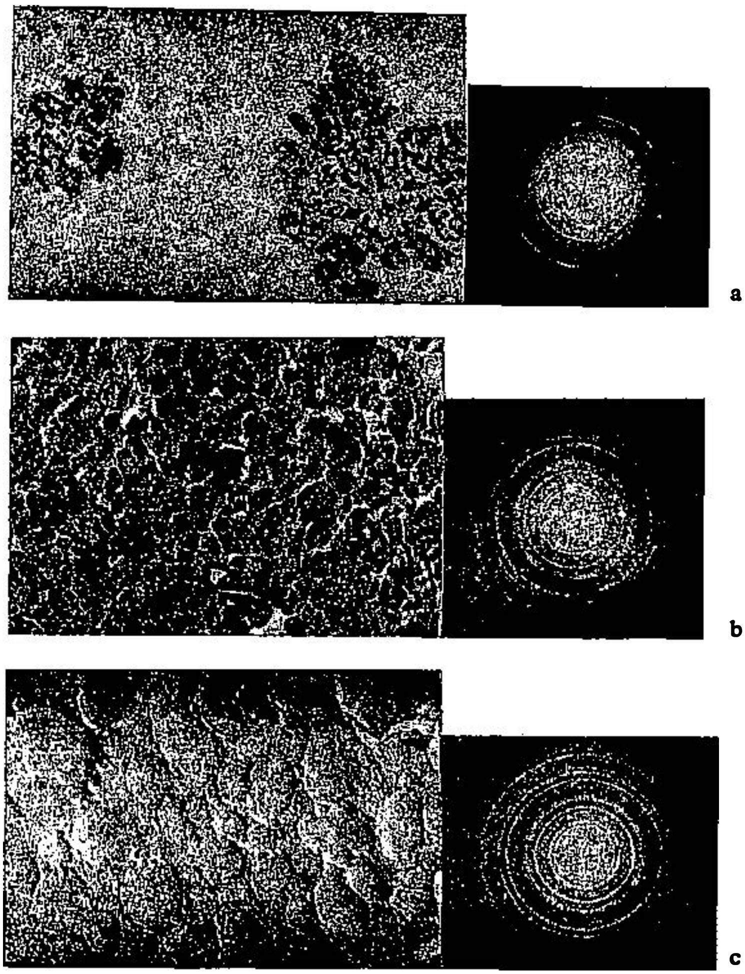
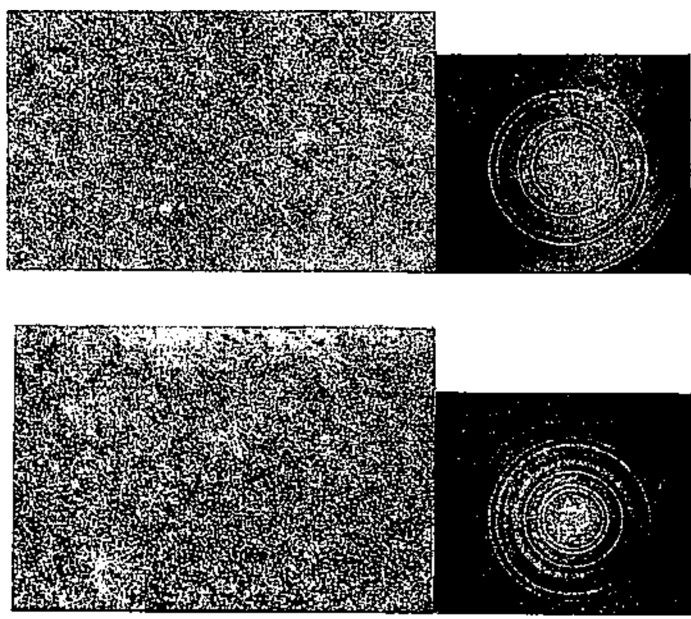


Fig. 1 a, b, c. A series of transmission electron micrographs and corresponding electron diffraction of lead sulphide thin films of different thicknesses on rock-salt substrate.

through initially deposided atoms taking up stable positions where they have lowest potential energy. The attainment of such a state is, however, dependent especially upon the rate of deposition and the thermal movement and the strength of forces between the deposit and the substrate alone[12]. Thin film having thickness 80 nm shows strongly oriented layer of lead sulphide, the particles have well defined geometrical shape with strongly oriented transmission electron diffraction which consist of arcs on rings having orientation (111) relative to substrate (Fig. 1 c). Micrograph shows that the film is uniformly thick over the whole area.

Film deposited on amorphous substrate consisted of randomly oriented polycrystals[2]. This was confirmed well from the series of micrographs (Fig. 2) which show the growth of lead sulphide on carbon film. The first stage in the growth process is the archipelago in which the film appears as a collection of widely separated islands (Fig. 2 a), followed by a labyrinth stage[13] in which the coalesced area are taking geometrical shape (Fig. 2 b). The transmission electron diffraction shows a polycrystalline structure which is normally on amorphous carbon.

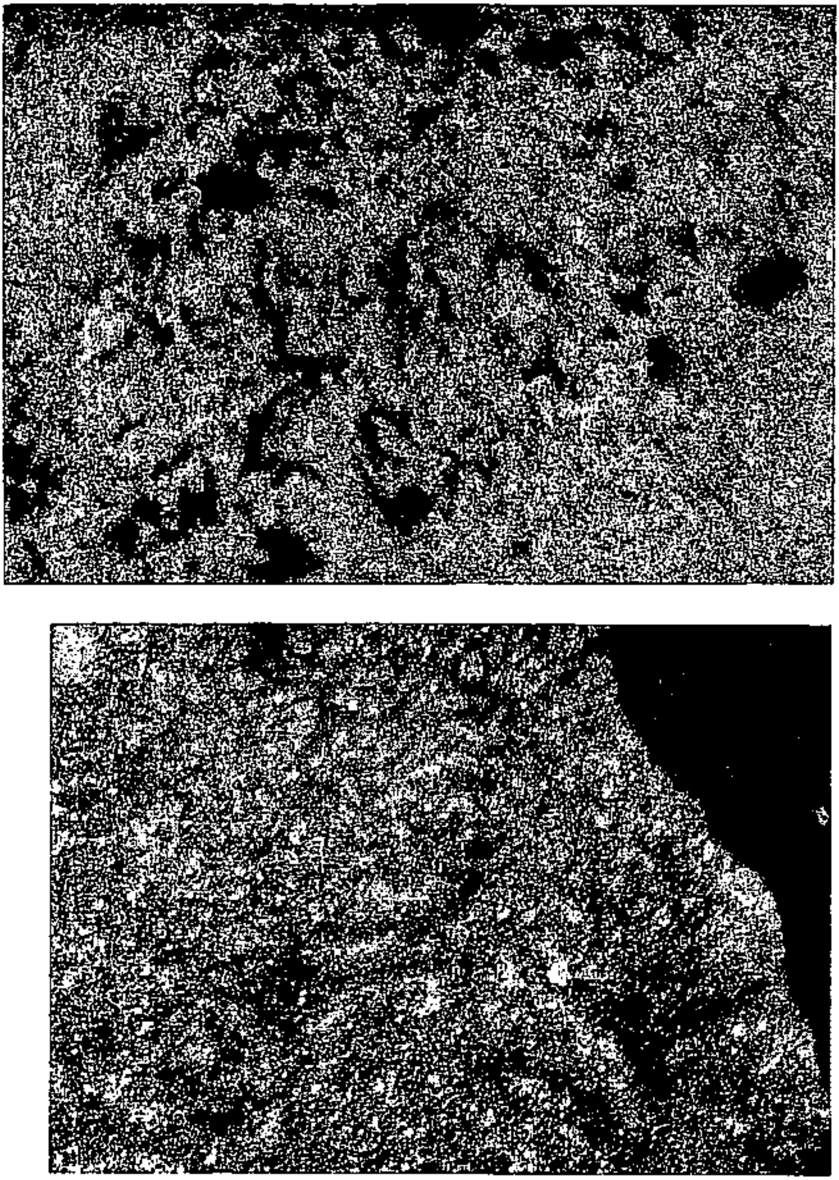


a

b

Fig. 2 a, b. A series of transmission electron micrographs and corresponding electron diffraction of lead sulphide thin films of different thicknesses on amorphous carbon film substrate.

Effeft of ionizing radiation on the structure of semiconductor (PbS) has been studied. The improvement of the crystalline structure during irradiation may be the first stage which can be explained by diffusion towards the surface of the non-stoichiometric atoms which exist in a free state among crystallites on dislocations, microfissures, etc. The diffusion of these atoms and the cleaning of the film from the non-stoichiometric atoms is considerably favoured during irradiation[14, 15]. When migrating through the sample under the effect of radiation the non-stoichiometric atoms may fill the vacancies of the crystalline lattice thus leading the improvement of its structure as shown in Fig. 3 a. At high irradiation structural defects begin to appear, characterized by a displacement of the atoms from the lattice into interstitial positions, which lead to the formation of Frenkel pair (Pb atom interstitial and S^{+2} ion vacancy). The mobility of Pb vacancies are more probable leading to the formation of voids and recrystallization of Pb resulting in the formation of different contrast areas (Fig. 3 b).



a

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Fig. 3 a, b. Transmission electron micrographs of lead sulphide thin films before and after irradiation with electron beam.

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ORIJENTACIJA I RAST TANKIH SLOJEVA OLOVNOG SULFIDA

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Rast i orijentacija tankih slojeva olovnog sulfida, naparenog na amorfne i kristalne podloge pri 300 K, proučavani su transmisionom elektronskom mikroskopijom i difrakcijom. Slojevi napareni na amorfnoj podlozi bili su polikristalni, dok su slojevi napareni na kalanoj NaCl podlozi bili orijentirani tako da je (111) PbS ∥ (100) NaCl. Proučavan je i utjecaj djelovanja elektronskog snopa na tanke slojeve PbS.