

## **X-ray studies on growth, thermal vibrations and internal stress in thin condensed films of Indium**

G S Narayana and N K Misra

Department of Physics and Meteorology, Indian Institute of Technology,  
Kharagpur-721 302, India

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Thin solid films prepared by evaporation technique are expected to be highly disordered. Large internal stresses and frozen-in lattice defects may develop during atomic deposition process (Kato *et al* 1961, Weiss and Smith 1962, Horikoshi *et al* 1962, Horikoshi and Tamura 1963, Klokholm and Berry 1968, Sen *et al* 1973, Nandi and Sengupta 1979). These will influence the Debye-characteristic temperature and structure-dependent properties of films. The study of these two physical parameters is of considerable interest in view of the applications of thin films in the technology of microelectronics. The X-ray diffraction technique provides an important tool for determining the Debye-characteristic temperature from the measurement of integrated intensities of the reflection profiles (Narayana and Misra 1988, 1989a,b) and internal stresses from the measurement of changes in lattice parameters (Keith 1956, Freedman 1962, Palatnik and Il'yinskii 1962, Kinbara and Haraki 1965, Vook and Witt 1965a,b, Haruta and Spencer 1966, Narayana and Misra 1989a,b,c). Narayana and Misra (1988, 1989a,b,c) have studied the thickness dependence of Debye-characteristic temperature and internal stresses in Pb, Sn and Ag films deposited on glass and mica substrates. Vook *et al* (1967) deposited thin films of Indium at  $-185^{\circ}\text{C}$  and studied the texture in the 100-1000 Å thick range and observed strong fibre texture (Vook 1975). The present investigation has been undertaken to make a detailed study of texture, to determine root-mean-square displacements of atoms in lattice points and internal stresses in thin Indium films of different thicknesses prepared by vacuum evaporation technique on glass substrates by X-ray diffraction method.

Preparation of the films, measurement of film thickness, recording of X-ray diffraction profiles, determination of integrated intensity and measurement of peak positions are carried out following the procedures described earlier by the authors (Narayana and Misra 1988, 1989a, b, c).

X-ray diffraction patterns from all these films were recorded using Cu-target. A strong  $\langle 101 \rangle$  texture developed for all the films. Relative intensities ( $I/I_0$ ) for different reflections are shown in the Table 1.

**Table 1.** Relative intensities ( $I/I_0$ ) for different reflections.

Film thickness (nm)	$I/I_0$				
	101	002	112	103	202
75	100	2.5	—	—	4.2
100	100	0.4	—	—	5.5
125	100	0.9	—	—	4.5
150	100	6.4	—	—	3.6
180	100	0.9	—	—	5.3
215	100	1.7	—	—	5.2
Thicker film	100	1.0	0.2	0.3	5.5
Bulk <sup>+</sup>	100	21	24	16	11

<sup>+</sup> ASTM data.

It appears from the Table 1 that a strong  $\langle 101 \rangle$  texture develops for all these films with a weak appearance of (002) plane parallel to the surface of the film. For thick films, polycrystallinity is observed, but strong  $\langle 101 \rangle$  texture is very much predominant.

The Debye-Waller factor  $B$  and root mean square amplitudes of atomic displacements along  $\langle 101 \rangle$  in different films were calculated following the equations (1) and (4) of Narayana and Misra (1988). Figure 1 shows the linear plot of  $\ln(I/AmLPF^2)$  vs  $2 \left( \frac{\sin \theta}{\lambda} \right)^2$  for films of different thicknesses along  $\langle 101 \rangle$ . The values of  $B$  and  $\langle \bar{u}^2 \rangle^{1/2}$  are shown in the Table 2.

Average  $d_{101}$  values are calculated for the films considering both  $\langle 101 \rangle$  and  $\langle 202 \rangle$  reflections using  $\text{CuK}_{\alpha 1}$  and  $\text{CuK}_{\alpha 2}$  radiations. Since  $d_{101}$  planes are parallel to the film surface, any change in  $d_{101}$  values will produce strain in the film perpendicular to the film surface. Therefore, an average internal stress perpendicular to the film surface is calculated from the following equation :

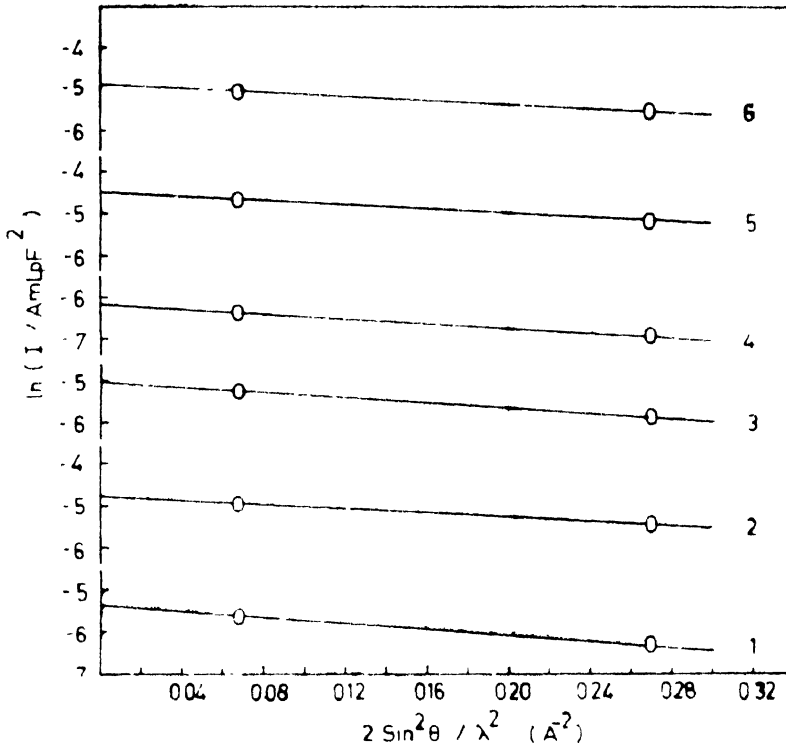
$$S = \frac{E_F}{2\nu_F} \frac{d_0 - d_F}{d_0} \quad (1)$$

where

$d_0$  is the  $d_{101}$  for bulk sample,

$d_F$  is the  $d_{101}$  for films,

$E_F$ ,  $\nu_F$  are Young's modulus and Poisson's ratio respectively for the deposits.



**Figure 1.** Plot of  $\ln(I / A mL_p F^2)$  vs  $2 \sin^2 \theta / \lambda^2$  for films of various thicknesses-(1) 75 nm, (2) 100 nm, (3) 125 nm, (4) 150 nm, (5) 180 nm and (6) 215 nm.

In the present calculations  $E_f$  and  $\nu_f$  are taken to be those of bulk material. Here  $E = 1.06 \times 10^{11}$  dynes/cm<sup>2</sup> and  $\nu = 0.45$  (Brandes 1983).  $d_{101}$  and  $S$  for films are shown in Table 2.

**Table 2.** Debye-Waller factor ( $B$ ), r.m.s. amplitude  $\langle u^2 \rangle^{1/2}$ , interplanar spacing  $d_{101}$  and internal stress  $S$  perpendicular to film surface.

Film thickness (nm)	$B$ (Å <sup>2</sup> )	$\langle u^2 \rangle^{1/2}$ (Å)	$d_{101}$ (Å)	$S (\times 10^9)$ dynes/cm <sup>2</sup>
75	3.70	0.217	2.717	0.87
100	2.67	0.184	2.717	0.87
125	2.86	0.190	2.716	0.43
150	2.55	0.180	2.717	0.87
180	2.27	0.170	2.716	0.43
215	2.03	0.160	2.716	0.43
Bulk†	---	---	2.715	---

† ASTM value.

It is observed from the Table 2 that the interplanar spacing  $d_{101}$  of the films are always larger than that of the bulk material. Similar results were also obtained by Vook *et al* (1967), Vook and Otooni (1968) and Narayana and Misra (1988, 1989 a, b, c) for tin, gold and lead films respectively on glass substrate. As the film thickness decreases, the dimension of the object decreases and the loosening of the atomic structure increases. One would expect larger amplitudes of atomic vibrations at the surface of the material than in its interior. These higher vibrational modes would result in an increased average interplanar spacing. This is obvious from the values of r.m.s. displacements  $\langle \bar{u}^2 \rangle^{1/2}$  along  $\langle 101 \rangle$  in Table 2. As film thickness increases,  $\langle \bar{u}^2 \rangle^{1/2}$  decreases. The stress developed perpendicular to the film surface is tensile but of negligible value. The variation of  $d_{101}$  and  $S$  with film thickness has been shown in the Figure 2. Toxen (1961)

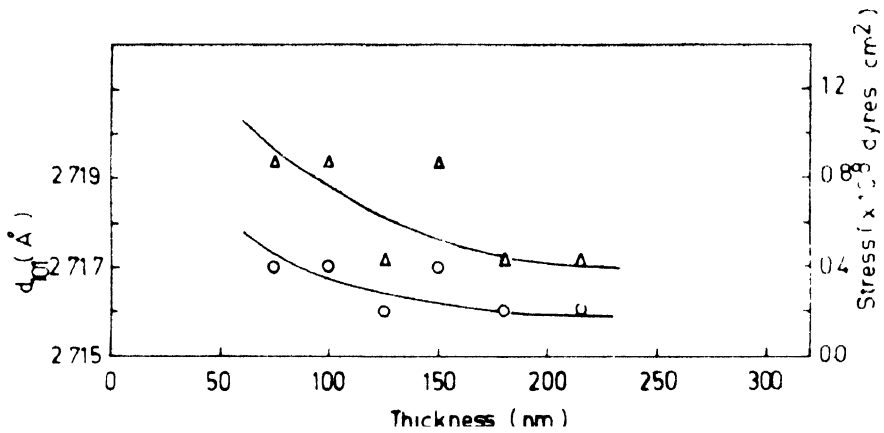


Figure 2. Plot of variation of lattice parameter  $d_{101}$  and internal stress ( $S$ ) with thickness of the films ( $\circ$  -  $d_{101}$ ,  $\Delta \Delta \Delta$  -  $S$ )

calculated stress in thin Indium films from the observed increase of their superconducting transition temperature with decreasing film thickness. The calculation was done on the assumption that the change in transition temperature is due to stress which has a maximum observable limited by the critical shear stress necessary to move dislocations. In this calculation stress was found to be quite large for very thin film (thickness  $< 200$  Å) but very small for films of larger thickness. Because of self-annealing of Indium films, intrinsic stress due to frozen-in defects is not expected.

Therefore, it can be concluded that thin Indium films with strong (101) texture can be deposited on glass substrate by vacuum evaporation method, that the measured increase in interplanar spacing is due to outward relaxation of the first few atom layers near the surface and that the internal stress developed is tensile in nature.

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