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RHEED INTENSITY OSCILLATIONS DURING THE GROWTH OF INDIUM ULTRATHIN FILMS

M. STRÓŻAK, M. JAŁOCHOWSKI AND M. SUBOTOWICZ

Department of Experimental Physics, Institute of Physics
Maria Curie-Skłodowska University
Pl. M. Curie-Skłodowskiej 1, 20-031 Lublin, Poland

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The ultrathin high-purity single-crystal indium films with atomically flat surfaces and precisely known thicknesses in UHV conditions were prepared. These films were deposited on the Si(111)-(7×7) and Si(111)-(6×6)Au substrate cooled to temperatures up to 110 K. The growth of the indium films was studied by reflection high-energy electron diffraction (RHEED). Pronounced specular beam intensity oscillations are found. The consequences for the understanding of RHEED intensity oscillations and of the growth of ultrathin films are discussed. The amplitude of the RHEED specular beam intensity oscillations as a function of the polar angle and temperature substrate Si was measured.

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1. Introduction

Reflection high-energy electron diffraction (RHEED) is very useful technique for studying growth and surface analysis of thin epitaxial structures prepared by molecular beam epitaxy (MBE). In particular the technique RHEED intensity oscillations has been used to process control growth layers in MBE [1]. The phenomenon of RHEED intensity oscillations was first observed during homoepitaxial growth of the semiconductors [2, 3]. In recent works the oscillations have been observed also during the growth of ultrathin metallic films with different lattice structure [4–6]. Oscillations in the intensity of RHEED patterns, observed under normal growth conditions, can be explained by a two-dimensional nucleation-on-terraces growth mode.

The information obtained from RHEED patterns is of potentially very wide ranging [1]. One can obtain informations about steps, islands and deduce the surface structure. It is also possible to derive more quantitative information about the surface structure such as positions of the surface atoms.

In this work we report the results of the RHEED intensity oscillations study during growth of the metallic ultrathin indium films deposited on Si(111) surface. Diffracted electron beam intensity was analysed as a function of the diffraction parameters (azimuth, incidence angle and different substrate structures). The temperature dependence of the substrate on RHEED intensity oscillations was discussed. We show that the growth mode of In on the Si(111) surface can be modified by evaporation of the other elements. About 0.5 atomic percent of Ag remarkably prolonged the layer-by-layer growth of Pb [7]. The RHEED rocking curve of the Si(111) surfaces was also measured.

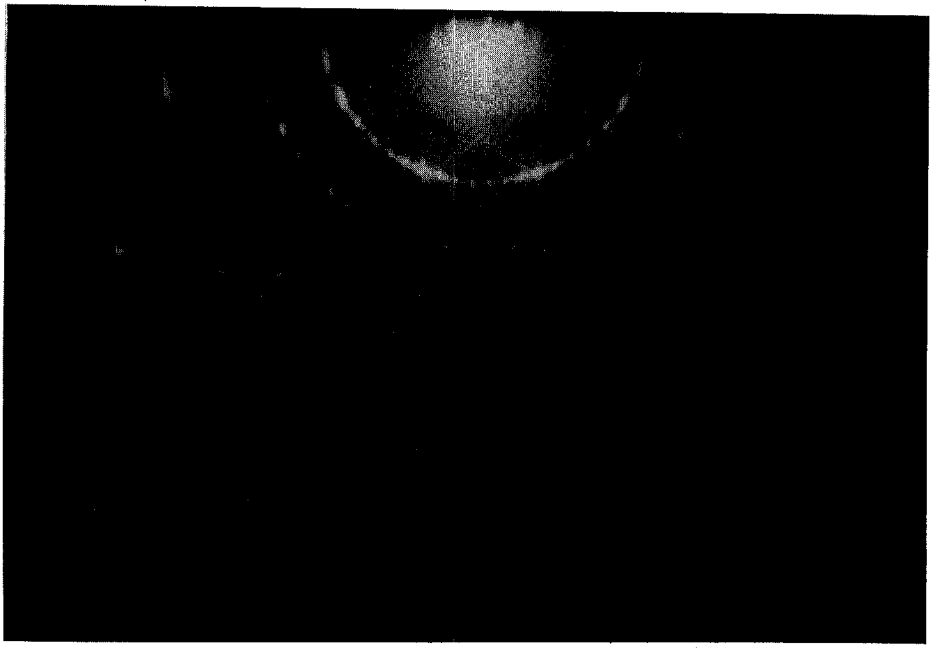
2. Experimental

The ultrathin In films were prepared in an ultrahigh vacuum (UHV) system [8] pumped by an ion pump and Ti-sublimation pump with a liquid-nitrogen (LN)-cooled cold wall. The base pressure was better than 1×10^{-10} hPa and the pressure during deposition was kept below 4×10^{-10} hPa. The substrates were Si(111) wafers with about 6 Ω cm specific resistivity at room temperature and typical dimensions $18 \times 4 \times 0.6$ mm. They were etched in $19HNO_3 \times 1HF$, rinsed in distilled water and methanol. After chemical etching the final surface cleaning was performed in the vacuum system before deposition by flashing for a few seconds at about 1550 K. This treatment removed the thin oxide layer and epitaxial SiC and resulted in the appearance of a sharp (7×7) superstructure RHEED patterns [9]. Typical RHEED patterns taken from Si(111)-(7x7) surfaces with $[11\bar{2}]$ incidence are shown in Fig. 1a. Direct resistive heating of the Si crystal was employed. The substrate could be cooled to about 110 K by making thermal contact between the rotatable holder and a LN container.

Figure 1b shows another type of surface prepared by deposition at about 1.2 monolayer (ML) of Au on a Si surface with (7×7) superstructure. Au deposits were annealed for 0.5 min at about 950 K and then the temperature was gradually lowered to about 500 K during 3 min. This type of surface showed a well-developed (6×6)Au superstructure RHEED pattern [7, 10, 11].

Indium was evaporated from stainless-steel crucibles and Au was evaporated from BN crucibles shielded by a water-cooled wall. Deposition rates between 0.01 and 0.1 nm/s were used. The thickness of the growing films was measured with a quartz-crystal monitor converting the frequency change into a voltage signal.

The RHEED system was operated at 20 keV and consisted of a triode electron gun, a 1-mm-diam beam-defining aperture, a magnetic focusing lens, and sets of magnetic deflection coils controlling the polar angle Θ of incidence of the electron beam. This angle could be adjusted with an accuracy of $\pm 0.05°$. The azimuthal angle was adjusted relative to the Si RHEED pattern with an accuracy of $\pm 1°$. The intensity of the specularly reflected beam was measured with aid of photodiode with a 0.8 mm diameter aperture attached to the fluorescent screen. The data from the photodiode and the quartz-crystal monitor were recorded both on a X–Y recorder and stored in digital form by computer system (microcomputer PC-XT with IEEE-488 interface system).



a

b

Fig. 1. RHEED patterns (a) taken from Si(111)–(7×7) structure, azimuth of incidence $[11\bar{2}]$; (b) taken from Si(111)–(6×6)Au structure induced by Au. Electron beam energy is 20 keV, glancing angle is equal to 0.35°.

3. Results and discussion

We observed RHEED specular beam oscillations for ultrathin metallic films of In during the growth on Si(111)-(7×7) Si(111)-(6×6)Au substrates. A typical result for a RHEED specular beam intensity measured during the growth of In films on Si(111)-(7×7) and Si(111)-(6×6)Au surfaces at 110 K is shown in Fig. 2. The polar (glancing) angle Θ of the electron beam was equal to 0.30° and azimuthal

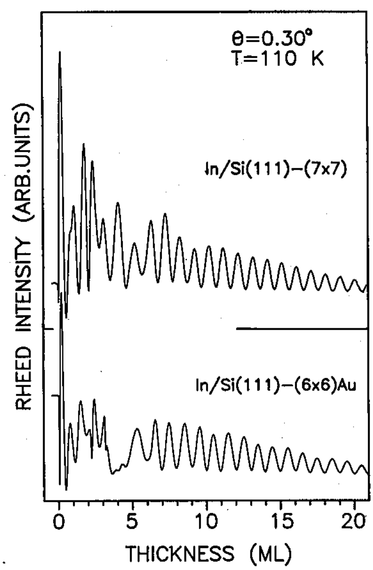


Fig. 2. RHEED specular beam intensity oscillations during the growth of In films on a clean Si(111)-(7×7) surface and on Si(111)-(6×6)Au surfaces at 110 K. Electron beam energy is 20 keV, azimuth: Si[11$\bar{2}$], polar (glancing) angle $\Theta = 0.30°$.

angle was chosen to be Si[11$\bar{2}$]. During the growth of the thin films on the pure Si(111)-(7×7) surface the oscillations became regular after the deposition of about 4 ML of In (about 10 Å). This initial complicated structure of the RHEED intensity oscillations is presumably caused by differences of the crystal structure of In and Si. The irregularity of the oscillations in the initial stage of the growth have been also created by the interaction of the beam with Si crystalline substrate and the growing films. This suggests that In up to 4 ML thickness on clean Si(111) forms a highly disordered films and strongly compressed layer in the same orientation as the thicker layers which seem to grow from the thin layer by recrystallization. Then sharp streaks appear. This is illustrated in Fig. 3 which shows the RHEED patterns of In layers on Si(111)-(7×7) surfaces after deposition 4 ML. For 2 ML of pure In films deposited on Si(111)-(7×7) surface the diffuse streaks in a strong background were not observed. The orientation of the In layers was the same as that of the substrate, In(111) || Si(111), for the thin films deposited directly on Si(111)-(7×7). When In grows on Si(111)-(6×6)Au surfaces, the regular oscillations are seen after

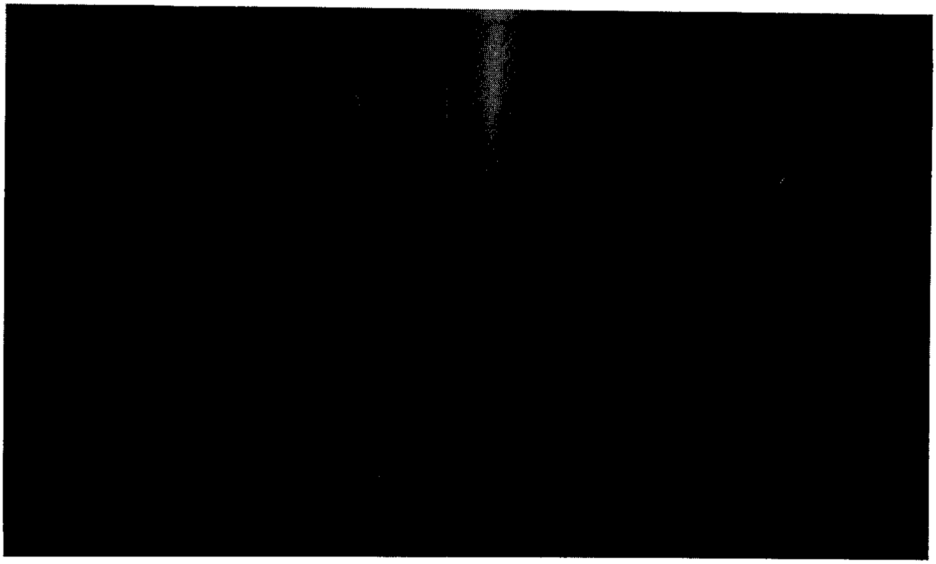


Fig. 3. RHEED patterns after deposition of the first 4 ML of In on Si(111)-(7×7) surfaces at 110 K. Electron beam energy is 20 keV, azimuth: Si[11$\bar{2}$].

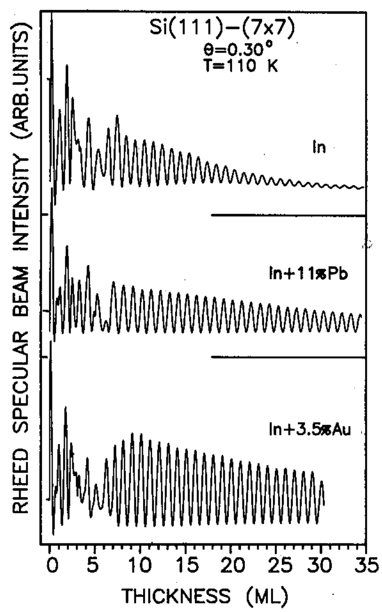
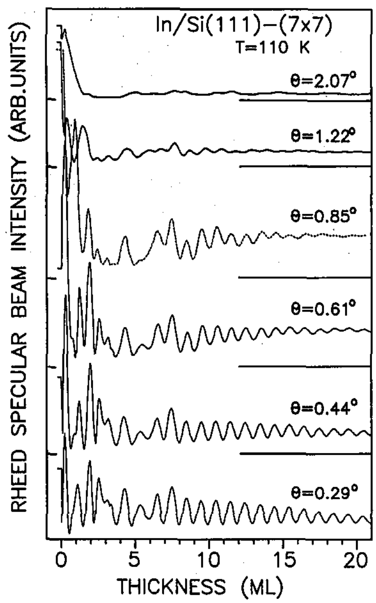


Fig. 4. RHEED specular beam intensity oscillations during growth of In+3.5%Au and In+11%Pb alloy films on a clean Si(111)-(7×7) surface at 110 K. Azimuth: Si[11$\bar{2}$], polar (glancing) angle $\Theta = 0.30°$.

deposition of about 6 ML of In.

For the In ultrathin films the intensity oscillations are strongly damped. Coevaporation of small amounts of Au or Pb enhanced strongly the RHEED intensity oscillations and the oscillations were observed over a large thickness range. This is shown in Fig. 4 in which Au and Pb was deposited simultaneously with In onto a Si(111)-(7×7) surface. More than 100 periods of oscillations were recorded for In+3.5%Au alloy films. This corresponds to a thickness of about 30 nm.



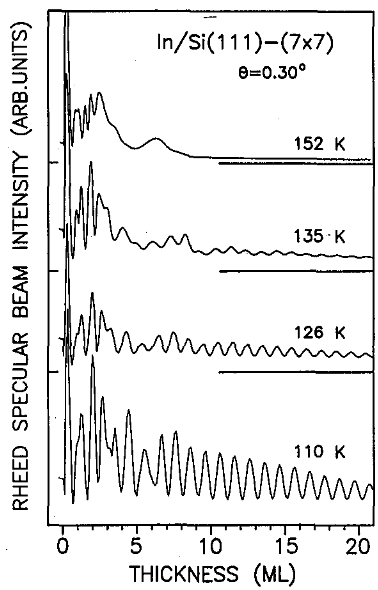


Fig. 5. RHEED intensity oscillations during the growth of In layers on a Si(111)-(7×7) surface at 110 K for the indicated polar angle. Azimuth angle of the incident beam direction was set at 7° from the [11$\bar{2}$] direction.

Fig. 6. RHEED specular beam intensity oscillations during the growth of In layers for different temperatures on clean Si(111)-(7×7) substrates. Azimuth: Si[11$\bar{2}$], electron beam energy 20 keV.

The RHEED specular beam intensity oscillations showed the strong glancing angle Θ dependence. Figure 5 shows the intensity of the specular beam as a function of the thickness of the In layer for different glancing angle of the incident beam. The RHEED intensities were measured at about 7° from the [11$\bar{2}$] direction of the incident azimuth. With increasing glancing angle the sharp modulated streaks are observed.

RHEED intensity oscillations occur not only during the growth at 110 K but also at higher temperatures. Figure 6 shows the RHEED intensity during the growth of the In layers on clean Si(111)-(7×7) surfaces at different temperatures. However the amplitudes decrease with increasing temperature and no oscillations were seen above 150 K. With increasing temperature the streaks become sharper. This suggests that we observed larger grain size growth of the indium films.

4. Conclusions

The results presented here should be of technological importance for the preparation of the ultrathin metallic films and for monitoring of their growth by means of the RHEED technique. We have shown that ultrathin single-crystal films of In with good surfaces can be grown on the Si(111) surface at about 110 K. The occurrence of the RHEED intensity oscillations during deposition of the In layers on Si(111)–(7×7) and Si(111)–(6×6)Au surfaces indicates monolayer-by-monolayer growth (Frank–van der Merwe (FM) growth mechanism). The manner of preparation of In thin layers has a remarkable influence. By coevaporation of the other elements (Au or Pb) the thickness range of the monolayer-by-monolayer growth of In films can be considerably extended.

Acknowledgements

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