Chapter 6

Development of a beam line and UHV Surface physics experimental facilities — Some results on Pb/Si(111)

Development of a beam line coupled with a ultrahigh vacuum (UHV) surface physics experimental facility with standard techniques like low energy electron diffraction (LEED), Auger electron spectroscopy (AES), sputter cleaning of the surfaces, and ultrathin film deposition is reported in this chapter. This facility allows in-situ ion scattering experiments on ultrathin films grown under UHV conditions. This is the first experimental facility of its kind in India. Having described the facility development, an example of a thin Pb layer deposition on a Si(111) surface and various analyses will be presented.

6.1 Introduction

At a fundamental level surfaces are of great importance because they represent a rather special kind of defect in the solid state. Much of our understanding of solids is based on the fact that they are, in essence, perfectly periodic in three dimensions. A large number of properties of a solid can be described in great detail using methods which rely on this periodicity. The introduction of a surface breaks this periodicity in one direction. This can
Development of Surface physics beam line

lead to structural changes on the surface and consequently many properties of the surface become different from those of the bulk solid.

Gaining a proper understanding of the surfaces is not only of academic interest, as there is growing interest in the properties of low dimensional structures in semiconductor devices, and a free surface can represent the simplest case of a two dimensional structure.

Conventionally epitaxial layers on single crystal surfaces are grown under ultrahigh vacuum (UHV) conditions in order to avoid contamination. Under atmospheric or non-UHV conditions usually an oxide layer is easily formed on semiconductor surfaces. These oxide layers hinder epitaxial growth [1-3]. The oxide growth can be inhibited in some cases by passivation of the substrates via chemisorption of different elements. For example, the oxide growth on Si(111) substrates can be hindered by passivating the Si dangling bonds with Br by a chemical method [3-5]. This passivated surface has been used for metallic film deposition on them under high vacuum conditions as in the studies presented in chapters 4 and 5. In these cases growth of epitaxial layers [6] and growth of micron sized epitaxial metal-silicide islands, e.g., gold-silicide islands, (triangular, trapezoidal, long wire like epitaxial structures) have been observed [7-9]. However, these systems are contaminated to some extent. For studies on atomically clean surfaces, we have developed a UHV experimental set up involving sample cleaning and preparation of atomically clean surfaces, monolayer level thin film(s) growth and studying various surface properties using diffraction of low energy electrons and Auger electron spectroscopy. The UHV set-up is connected with one of the beams (−30°) of the 3 MV tandem accelerator facility to carry out ion scattering (RBS/ channeling) measurements with MeV ion beams. The beam line and the details of the experimental set-up are described in the following section. Some experimental results involving growth and characterization of Pb on Si(111) surfaces are presented in section 6.3.

6.2 General description of the surface physics beam line and the experimental facilities

The schematics of the surface physics beamline is shown in Fig. 6.1. The various positions of the focusing (magnetic quadrupole doublet lens) and beam diagnosing (beam profile monitor, Faraday cup etc.) elements and the target location have been decided using the
ray tracing code [10] developed at IOP. In order to get a parallel ion beam of about a 1 mm diameter for the ion channeling measurements, two collimators separated by a distance of about a meter are placed just before the UHV chamber. These collimators also help in maintaining a differential pressure (i) from the UHV chamber to the intermediate section pumped by a turbo pump and (ii) from the intermediate section to the main beamline, during the experiments. The pressure in the beamline is \( \sim 10^{-7} - 10^{-8} \text{ mbar} \). The drift tubes for the beam line, beam view port and slits were fabricated at SMP Pune BPM, Faraday cup and quadrupole doublet lens were procured from National Electrostatics Corporation in the U.S.A.

Fig. 6.2 shows two photographs of the UHV chamber along with various components. The UHV experimental facility is provided with sample cleaning with Ar\(^+\) ion sputtering, thin film(s) growth by deposition from Knudsen cell(s), low energy electron diffraction (LEED)/Auger electron spectroscopy (AES) measurements using a reverse view SPECTRALEED set up and ion scattering (RBS/channeling) measurements using an MeV ion beam from the tandem ion accelerator. For the growth of multi-elemental thin films three Knudsen cells are mounted on a water cooled cluster flange assembly. The Knudsen cells consist of 5 cc capacity PBN crucibles with maximum evaporation temperature up to 1700 \(^{\circ}\)C. Each K-cell is provided with water cooling and independent shutter assembly. The cells are aimed at the sample in such a direction that to have an equal coverage of the thin films while depositing on the sample. A load-lock system with sample heating facilities has been attached with the UHV chamber for the initial sample degassing and sample transfer. The sample transfer from the load lock chamber to the main chamber is done with the help of a magnetically coupled transfer rod and loading/unloading fork. The load-lock chamber is provided with a fast entry hinged flange for quick loading/unloading of the samples and a manually controlled leak valve for venting the system with dry nitrogen gas, when required. In the main UHV chamber the sample positions are manipulated by a 6-axis (X, Y, Z, rotational, azimuthal, tilt motions) precision goniometer. An e\(^-\)-beam sample heating facility is provided with the goniometer. There is also a residual gas analyzer (RGA) attached with the UHV chamber in order to determine the various contaminations present inside the chamber. The equipments and their physical parameters of our surface physics experimental facility are listed in Table 6.1.
Fig. 6.1. Schematic diagram of the surface physics beam line and experimental facilities with the 3 MV tandem accelerator
Fig. 6.2. Photographs of the UHV chamber along with various components attached to it are shown in (a) and (b). A part of the beam line with various components is also seen in (b).
Table 6.1. Equipments and their physical parameters of the Surface physics experimental facility

<table>
<thead>
<tr>
<th>Equipment/Accessory</th>
<th>Description</th>
</tr>
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<tbody>
<tr>
<td>UHV chamber (10^{-10} mbar)</td>
<td>18 inch dia, 25 inch high cylindrical chamber, Starcell 400 l/s Ion pump + TSP</td>
</tr>
<tr>
<td>Load-lock chamber (10^{-8} mbar), (High Vacuum Process, Italy)</td>
<td>12 inch dia, spherical chamber, 500 l/s Varian Turbo pump, 3 target (1 inch dia, Molybdenum) stations, resistive heating, Max 800 °C (±1 °C), 80 cm travel, bayonet type sample transfer</td>
</tr>
<tr>
<td>Indexer table with sample heating, magnetically coupled transfer rod</td>
<td></td>
</tr>
<tr>
<td>6-axis Gamometer, (Johnsen Ultravac, Canada)</td>
<td>X,Y=± 12.5 mm, Z=500 mm</td>
</tr>
<tr>
<td>sample heating with Eurotherm 2408 controller</td>
<td>Rotation (θ) &amp; Azim (φ) = 0 – 360°(0.1° prec), Tilt (ψ) = ±30° (0.1° prec)</td>
</tr>
<tr>
<td>Electron beam heating, Max 1200 °C (±1 °C)</td>
<td></td>
</tr>
<tr>
<td>Ar⁺ ion gun with electromagnet (AG 5000, VG Microtech, UK)</td>
<td>Energy 0 - 5 keV, mm spot size 1 cm dia</td>
</tr>
<tr>
<td>LEED/ AES (Omicron, Germany)</td>
<td>Reverse view SPECTRALEED optics, 4 grids, electron energy 0 - 3.5 keV, 98 mm retraction, Computer controlled AES</td>
</tr>
<tr>
<td>Knudsen Cells (Three), EK-100 Controller (High Vacuum Process, Italy)</td>
<td>On a cluster flange with cooling jacket, 5cc PBN crucible, independent shutter, cooling, Max evaporation temperature 1700 °C</td>
</tr>
<tr>
<td>Residual Gas Analyzer Prisma, QMS 200 (Balzers, Germany)</td>
<td>Mass 0 - 300, M/ΔM = 84, Faraday cup and Channeltron</td>
</tr>
<tr>
<td>RBS/channeling, irradiation with tandem accelerator RBS detector (area, scattering angle, solid angle)</td>
<td>He ion beam (1-3 MeV) and other ions with ME/q^2 ≤ 35 amu-MeV, SB (EG&amp;G, Ortec, USA), (50 mm², 160°, 2 mstr), Reso 12 keV @ 5.4 MeV Am^{241}</td>
</tr>
</tbody>
</table>
6.3 Some results

6.3.1 Preparation of a clean surface

Preparation of an atomically clean surface is a requirement for surface physics experiments. Sample surface is usually cleaned either by Shiraki etching [11] or by sputtering with inert gas ions. Auger electron spectroscopy is usually used to detect contaminations on the surface. Figure 6.3 shows the Auger electron spectra (AES) of a carbon-contaminated Si(111) sample, which was subsequently cleaned with Ar ion sputtering.

Fig. 6.3. Auger electron spectra (AES) from a carbon contaminated Si(111) sample, which was subsequently cleaned with 2.5 keV Ar ion sputtering.
6.3.2 Growth of Pb films on a Si(111) surface

6.3.2.1 Introduction

In recent years there has been significant interest in thin metal films on semiconductors from fundamental and technological points of view. From technological standpoint, these films are important in the fabrication of integrated circuits. There has been efforts to understand the formation of Schottky barriers on an atomic scale. Unreactive epitaxially grown metal-semiconductor contacts are considered ideal systems for this purpose. Pb/Si and Pb/Ge systems are prototypes of such unreactive metal-semiconductor contacts. Other aspects like order-disorder transition and melting of monolayer Pb on semiconductors, especially on Ge, have been the subject of many studies [12-15]. Such systems also show charge density wave transition [16]

In particular, Pb/Si(111) has been proposed as a model system because Pb and Si have negligible mutual bulk solubility [17,18]. The Pb/Si system is remarkably unreactive, and ought to provide a relatively simple interface to study. There has been numerous studies on formation of various structural phases in submonolayer and monolayer growth of Pb on different reconstructed Si surfaces [17,19-21]. In order to demonstrate the capability of our newly developed surface physics facility, here we have taken up a case study of the growth of Pb layers on a Si(111) surface.

6.3.2.2 Experiment

Chemically Br-passivated Si(111) (P doped with resistivity 10 – 20 Ω-cm) substrates, which we used in many earlier studies including those in chapter 4, were loaded on a six-axis manipulator in the UHV chamber. Traces of C impurity observed (seen in Fig 6.3) on the Br-Si(111) substrate were sputter cleaned with Ar⁺ ions from a gas of 99.9% purity. 2.5 keV Ar⁺ ions with a beam flux of ~ 1 x 10¹³ ions/cm²/sec were incident on the substrate at an oblique angle (40° to the sample surface normal) to enhance the sputtering. The sample was annealed subsequently to regain the crystalline surface. Some times the traces of impurities like C reappear after annealing, so the Ar sputter cleaning and subsequent annealing cycle is repeated till removal of these impurities beyond the detection level. However due to the malfunctioning of the sample heating system on the sample manipulator, the maximum temperature achieved was around 500 °C. Observation of enhancements in the Si surface peak intensity in the RBS spectrum under the ion channeling condition (spectra not shown...
here) indicate the presence of disordered layers in the surface region. Also no ordered LEED pattern was seen. So our experiment involves a clean Si(111) substrate but with a disordered surface layer (range of 2.5 keV Ar⁺ ions in Si in oblique incidence is \( \sim 45 \pm 16 \text{ Å} \)).

About 10 nm thin Pb (5N purity) films were deposited onto the Si(111) substrate at room temperature from one of the K-cells placed at a distance of 20 cm from the target. A deposition rate of 0.12 nm per min. was achieved by keeping the K-cell temperature at \( \sim 570° C \) at a base pressure of \( 9 \times 10^{-9} \text{ mbar} \). The rate of deposition was determined by calibrating the Pb thickness using RBS measurements (shown in Fig. 6.4). AES measurements were made at various time intervals during the Pb deposition. The Pb/Si(111) sample was subsequently annealed at 300°C for 30 min. RBS/channeling measurements are made on as-deposited as well as annealed samples. Morphology of the annealed sample was studied using atomic force microscopy (AFM).

![Fig. 6.4. Calibration of Pb deposition as a function of time using RBS measurements](image)

**Fig. 6.4.** Calibration of Pb deposition as a function of time using RBS measurements

### 6.3.2.3 Results

**A. Auger electron spectroscopy**

Auger electron spectroscopy can be used to study the growth mechanism of Pb on Si(111)
Fig. 6.5 shows the AES signals from a Pb/Si(111) sample as a function of deposition time. Unfortunately the main Pb(NO), (MOO) (94 eV) and Si(LVV) (92 eV) Auger lines are too close. Although this complicates the analysis, we can still use AES to get some information about the growth mechanism. The spectra were recorded in the derivative mode and the intensity is determined by numerical integration. Usually this sort of analysis is performed following the evolution of the peak to peak height of the Auger intensity. However, here the measurement of the peak to peak heights will produce erroneous results because the line shape changes as the coverage increases.

In order to determine the growth modes, we have determined the areas of the peak of the first Pb line (90 eV) and the dip of the second Pb line (94 eV) in the AES spectra (seen in Fig. 6.5). These areas as a function of deposition time have been plotted in Fig. 6.6. The first dip and second peak of Pb is heavily influenced by the Auger signal (92 eV) from...
the Si substrate, so these were not taken into consideration.

\[ \text{AES peak area (arbitrary unit)} \]

\[ \begin{align*}
\text{deposition time (sec)} & \\
0 & 25 50 75 100 125 150 175 200 225 \\
\end{align*} \]

**Fig. 6.6.** Evolution of the AES intensities of the Pb lines during room temperature deposition of Pb onto the Si(111) surface. Areas of the first peak and the second dip are plotted as a function of time. Dashed lines are just guide lines indicating completion of one monolayer.

It is known that even at room temperature Pb on Si(111) grows according to the Stranski-Krastonov mechanism, i.e., first one monolayer is completed and subsequent growth proceeds in islands [17,21]. Therefore after the completion of the first monolayer (1ML = 7.8 × 10^{14} atoms on Si(111) surface), a kink appears on the AES intensity evolution plot. One ML of Pb atoms on Si(111) surface corresponds to about 2.86 Å in thickness [17] (the bulk Pb-Pb distance is 3.50 Å). From the RBS calibration of the Pb film deposition rate (Fig 6.4) we have estimated the completion of the first ML will occur after \( \sim 145 \) sec deposition time (shown in Fig 6.6). We have studied thick (\( \sim 35 \) ML) Pb layers to explore their crystallinity and morphology aspects. These are described below.
B. Pb-induced ordering of disordered interface Si

RBS/C measurements on the as-deposited and the 300 °C-annealed sample are shown in Fig 6.7. Pb seem to be polycrystalline with no improvements in the crystalline quality (Compared to random RBS there is no reduction in the Pb signal when the ion beam is aligned with [111] axis). We have not checked if there is reduction in yield for directions with small tilt with Si[111]). The spectra from the annealed sample shows the broadening of the Pb signal. This may be due to either the diffusion of the Pb into the Si substrate or formation of islands. But Pb/Si interface is reported to be abrupt [17]. So the broadening appears to be due to islanding, which is also confirmed by AFM. We have chosen an annealing temperature of 300 °C because it is below the Pb melting point (328 °C), and desorption of the Pb is negligible at this temperature [17,22].

Although there is no change of crystalline quality of Pb upon annealing, there is some improvement in the crystalline quality of Si in the interface region as seen in the aligned spectra of the Si. These indicate a Pb-induced recrystallization of the disordered Si(111) surfaces. In Si, $\chi_{\text{min}} = 19\%$ for the as-deposited samples comes down to 17% upon annealing. [The Br-passivated Si(111) sample, before any treatment had a $\chi_{\text{min}} = 4.7\%$.] This is due to reduced dechanneling at the Pb/Si interface region, indicating partial reordering of the disordered Si at the interface. Earlier studies of the behaviour of Pb on Si(111) surfaces have shown that Pb allows near-surface Si atoms to move at low temperatures. For example, 1 ML Pb on the (7 x 7) reconstructed Si(111) surface, after annealing to temperatures below 300 °C, is able to remove the stacking fault present in half of the (7 x 7) unit cell of the reconstructed Si(111) surface [23]. However, Pb-induced ordering of a disordered Si layer, observed in this work, has not been reported earlier. This Pb-induced ordering is possibly the reason why thick homoepitaxial Si layers can be grown by the mediation of 1 ML Pb at unusually low growth temperatures [24,25].

A schematic representation of the change in Si RBS/C yield (as observed in Fig 6.7) due to the Pb island thickness and lateral area coverage is shown in Fig 6.8. From a preliminary analysis of the Si edges of the RBS spectra, majority of the islands have been found to have a thickness of $\sim 35$ nm covering $\sim 25\%$ area, the remaining area on the average has a lower thickness ($\sim 1$ nm) of Pb.
Fig. 6.7. RBS/C spectra of the as-deposited and the 300°C-annealed sample obtained with a 2.0 MeV He\(^+\) beam. Note the energy (channel no.) scale. It is expanded in the Pb region.
Fig. 6.8. Schematic representation of the Si RBS/C yield from (a) bare Si (b) uniform layers of Pb/Si (c) Pb islands over a thin uniform layer on Si in layer-plus-island mode are shown.

C. Morphology of 300 °C-annealed Pb layers on Si(111) surface

AFM micrograph (Fig 6.9) shows the islanding of Pb in the case of 300 °C-annealed sample. Also smaller grains are seen. At 300 °C, high surface mobility is responsible for the agglomeration of the Pb atoms into large islands.
Fig. 6.9. AFM micrograph of the 300 °C annealed sample, showing small grains and growth of bigger Pb islands

Fig. 6.10 shows a high resolution AFM micrograph of a region containing Pb islands of various shapes. One can see most of the bigger islands have ~ 0.5 μm lateral dimension. Most of the islands tend to have the same shape. From the high resolution AFM micrograph shown in Fig. 6.10 the islands appear to be hexagonal, as found by Westerling et al. [21]. In Fig. 6.11 a cross-sectional analysis of the Pb film is shown.
Fig. 6.10. High resolution AFM micrograph of the 300 °C annealed sample shows islands with facets.
Fig. 6.11. Cross-sectional analysis of the annealed sample showing the exposed Si surface, possibly with a thin Stranski-Krastanov Pb layer on Si, and islanding in the Pb layer.

From the AFM micrographs presented above the lateral coverage of the thin uniform layers of Pb on the annealed sample does not appear to be 75% as determined from the RBS/C spectra. However if one considers also the Pb depleted regions between the grains, then the average coverage area of the thin uniform layer would increase. The RBS and AFM measurements were not made at the same spots on the sample. There may be some nonuniformity in the surface morphology.
6.4 Summary and outlook

A brief description of the development of the surface physics experimental facility with some results has been presented. Connection of the UHV facility with one of the beamlines (−30°) of the tandem accelerator has made the various surface and interface characterization capabilities much more versatile. As a demonstrative case, we have reported the growth of Pb on clean Si(111) surface. RBS/C results have provided the evidence of Pb-induced recrystallization of a disordered Si(111) surface.

The developed experimental facility can be used to study atomically clean bare surfaces of single crystals, which show interesting reconstructions and relaxations, and growth of metallic epilayers on them. Metallic single (multi) layer(s) (e.g., Ag, Au) on Si surfaces can be grown and characterized and compared with all results obtained under high vacuum growth on Br-passivated Si surfaces. Thermal behaviour including melting of confined layers, which is expected to be qualitatively different from bulk melting, can be studied. Structural transitions on surfaces and on thin layers can be studied. Also, the effects of high energy ion irradiation on the in-situ grown metallic thin films on Si can be studied and compared with similar irradiation studies on ex-situ metallic films grown on Br-passivated Si surfaces under high vacuum conditions [26], as those presented in chapter 5. Clustering phenomenon on surfaces [27] can be effectively studied with this facility. This is the first facility of its kind in India, combining standard surface science techniques with Rutherford backscattering spectrometry and channeling techniques under ultrahigh vacuum environment.
Bibliography


Development of Surface physics beam line


