Electron transport across complex oxide heterointerfaces
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Chapter 2

Experimental details

This chapter outlines the key techniques used for the growth and characterization of epitaxial as well as non-epitaxial thin films used in this thesis. First, the details of the surface preparation of the semiconducting substrate prior to the film deposition are explained. This is followed by a brief description of the steps followed for the fabrication of Hall bars, devices for BEEM studies and magnetic tunnel junctions. Next, the experimental techniques that have been employed to probe the macroscopic and nanoscale electronic transport properties across the hetero-interfaces as well as in thin films are explained. In the last part of the chapter, experimental details of magnetic tunnel junctions measurements are discussed.

2.1 Substrate surface preparation

SrTiO$_3$ (STO) is an ABO$_3$ type cubic perovskite. (001) oriented SrTiO$_3$ substrates consist of an alternating arrangement of non polar SrO and TiO$_2$ planes along the c-axis, as explained in chapter 1. For SrTiO$_3$ (001), the out-of-plane lattice constant, $d$, is defined as the distance between two consecutive identical planes along the c-axis and is equal to 3.905 Å. The surface terminating plane of SrTiO$_3$ determines the atomic layer sequence of the epitaxially deposited films. As electronic transport in our devices strongly depends on the structural quality of the heterointerface, the preparation of a high quality singly terminated substrate is a prerequisite. Since TiO$_2$ termination of SrTiO$_3$ surface is predicted to be energetically more stable than SrO termination [1], [2], we have used TiO$_2$ terminated Nb:SrTiO$_3$ substrates in order to obtain well controlled interfaces for our experiments. To achieve a clean, atomically flat, TiO$_2$ termination, Nb:SrTiO$_3$ is chemically treated and thermally annealed. Thin films of Au or Cu or La$_{0.67}$Sr$_{0.33}$MnO$_3$ are grown on a singly terminated Nb:SrTiO$_3$ substrate whereas the magnetic multilayers are grown on singly terminated SrTiO$_3$.

Chemically and mechanically polished substrates, with a miscut angle $<0.5^\circ$, obtained from the crystal manufacturers, Furruchi, Japan and Crystec, GmbH, Germany, have a mixture of SrO and TiO$_2$ termination. We use a well established protocol from earlier work by Kawasaki et al., and G. Koster et al., for achieving single
2. Experimental details

Figure 2.1: AFM images for 0.01 wt% Nb doped SrTiO$_3$ after chemical treatment (a) and after annealing (b). (c) Line cross section of AFM image after annealing, which shows a step height = 4 Å.

terminated surfaces [3], [4]. Ultrasonication in de-ionized (DI) water for 30 minutes enhances the etch selectivity of the more base layer of SrO with respect to TiO$_2$ by its hydroxylation i.e., the formation of Sr(OH)$_2$.

\[
SrO + H_2O \rightarrow Sr(OH)_2
\]  

Sr(OH)$_2$ can be removed by buffered hydrofluoric acid in 30 seconds and this results in a TiO$_2$-terminated surface.

\[
Sr(OH)_2 + BHF \rightarrow Sr^{2+} + 4F^- + 2H_3O^-
\]  

As a final cleaning step, these substrates are ultrasonicated in DI water for another 20 minutes. After this chemical treatment, the substrates are annealed at 960°C in an O$_2$ flow of 300 cc/min to yield an atomically flat and straight terraced surface.
2.2 Thin film growth

The annealing time depends upon the width of the terrace; the higher the width, longer is the time required for annealing. For all the work reported in this thesis, the substrates are TiO$_2$ terminated using the above procedure. Figure 2.1 shows the atomic force microscopic images of Nb doped SrTiO$_3$ substrates after chemical treatment (a) and after thermal annealing (b). Figure 2.1 (c) shows the line cross section of the AFM image taken after annealing, with a step height corresponding to 4 Å (step height of unit cell of TiO$_2$ sub-lattice)

2.2 Thin film growth

We have grown thin films of La$_{0.67}$Sr$_{0.33}$MnO$_3$ (100) (LSMO) on SrTiO$_3$ (100) as well as on Nb: SrTiO$_3$ (100) single crystal substrates. Multilayers are also grown for investigating tunnel spin polarization of LSMO films in magnetic tunnel junctions. The technique of Pulsed Laser Deposition (PLD) is employed for the epitaxial growth of thin films of LSMO, where the growth is monitored using in situ reflection high energy electron diffraction (RHEED).

2.2.1 Pulsed laser deposition and RHEED

Pulsed Laser Deposition (PLD) is a well known technique for growing stoichiometric complex oxides in an oxygen environment. In PLD, a high power pulsed laser is focused on a target kept in an oxygen environment. The strong absorption of the electromagnetic radiation by the solid surface leads to a rapid evaporation of the target material. The plasma plume deposits on the substrate lying at distance of 40 mm to 60 mm from the target. We use the PLD system (obtained from TSST) in the lab of Prof. B. Noheda at the University of Groningen. A KrF excimer laser (Lambda Physik COMPex Pro 205) with a wavelength of 248 nm and a pulse duration of 25 ns is employed in the PLD set up. Using a specially designed mask of 23×7 mm$^2$, the most homogeneous part of the laser beam is selected. This mask design comprises of multiple rectangular holes whose image on the target is used to obtain the sharpest focus of the laser. Figure 2.2 (a) shows the schematic of a PLD setup. The high energy pulsed laser irradiation causes a congruent evaporation of the target to form a plasma plume. Such a congruent evaporation caused by a rapid heating rate helps in stoichiometric transfer of the material from the target onto the substrate [5]. Figure 2.2 (b) shows a plasma plume during a deposition in an oxygen pressure of 0.35 mBar. An optimum oxygen pressure, target to substrate distance ($d_{TS}$), laser energy per unit target area (laser fluence), and the substrate temperature are the growth parameters which play an important role in obtaining highly crystalline and stoichiometric films. LSMO films are deposited in an oxygen
pressure of 0.35 mBar with a laser fluence of 2 Jcm$^{-2}$ (1 Hz) on SrTiO$_3$ or Nb:SrTiO$_3$ substrates heated to 750$^\circ$C. We use the laser spot size of 2.2 mm$^2$ and a $d_{TS}$ of 53 mm for epitaxial growth of LSMO films (details in chapter 4). For obtaining a homogeneous ablation, the target is continuously scanned during the deposition. After growth, the LSMO films are cooled down to room temperature in the presence of oxygen at a pressure of 100 mBar. The multiple target holder allows us to grow in situ multilayer structures such as magnetic tunnel junctions and spin valves.

Growth of thin films can be monitored at the angstrom level using in situ reflective high energy electron diffraction (RHEED). In RHEED, an electron beam of 30 keV hits the surface of the crystalline substrate at a grazing angle (less than 3$^\circ$). The grazing angle ensures surface sensitivity and hence is used as a tool to monitor the deposition of thin films during growth [6]. It is common to use RHEED for low pressure growth of epitaxial films. To achieve the high oxygen pressure required for the growth of complex oxide thin films, a differential pressure is created in the chamber and inside the RHEED tube in order to minimize the electron scattering by oxygen atoms. Such a state of the art technique was developed in the group of Prof. G. Rijnders at the University of Twente. Most of the incident electrons are elastically scattered by the first few atomic planes below the surface of the sample and a diffraction pattern is formed on the phosphorescent screen. The reciprocal space associated with the 2D lattice of a perfect crystal surface consists of infinite rods perpendicular to the surface of the sample and passing through the 2D lattice points. A RHEED pattern will then be determined by the intersection of these rods.
2.2. Thin film growth

Figure 2.3: (a) A Schematic of the formation of RHEED spots on a phosphorus screen. (b) Typical RHEED specular spots along with the direct spot image captured using a camera.

with the Ewald sphere, whose radius is inversely proportional to the wavelength of the incoming electron beam. If the surface is perfectly flat, the reciprocal rods have no width and this intersection gives rise to a pattern of spots aligned along semicircles (Laue circles), as schematically shown in Fig. 2.3 (a) combined with a schematic of elastically scattered electrons in real space. If the few electrons penetrate into the substrate, the reciprocal rods are modulated, with a periodicity of the c-axis parameter of the substrate. The intensities of the spots of the diffraction pattern change according to the surface coverage and roughness [7]. We use these changes to follow the thin film growth. A maxima in the intensity is obtained for a full monolayer and hence the number of monolayers deposited are monitored during deposition. Figure 2.3 (b) and (c) show RHEED spots before and after the deposition of 10 nm LSMO films on Nb doped SrTiO$_3$ substrate. In the first part of the work presented in this thesis, ultra thin (a couple of nm) films of LSMO are deposited using PLD in combination with RHEED for exploring their electronic transport properties. In the remaining part, thicker films of LSMO as well as multilayered structures are grown and investigated for their spintronic properties. SrTiO$_3$ has a lattice parameter of 3.905 Å and LSMO grows epitaxially on SrTiO$_3$ with a lattice mismatch < 0.3%.

2.2.2 Electron beam evaporator

We have grown thin films of Au and Cu in order to study the electronic transport across their interface with Nb:SrTiO$_3$. Nb doped SrTiO$_3$ is used as a hot electron collector (semiconductor) in a transistor configuration i.e., emitter/base/collector,
2. Experimental details

For the work reported in this thesis; and hence its interface with well known metals was studied. For growing Au and Cu films on Nb: SrTiO\(_3\), we have used electron beam evaporation using a Temescal Thin Film Coater 2000 (TFC-2000). TFC-2000 has a bell jar shaped load lock which is separated from main chamber via a gate valve. Samples are placed in the load lock for deposition whereas the target is placed in the main chamber. The pressure of the chamber during deposition is 6\(\times\)10\(^{-7}\) mBar. In e-beam evaporation, a target is bombarded with an energetic electron beam in high vacuum, where the acceleration voltage is 10 kV. Eight different materials can be grown using TFC-2000. A quartz crystal microbalance is used to determine the deposition rate and final film thickness. We have used a growth rate of 1 Ås\(^{-1}\) to obtain thin films of Au and Cu, having a root mean square roughness of 0.4-0.6 nm, as observed using atomic force microscopy (AFM). The thickness of the films is monitored using a quartz crystal oscillator during the growth. We used e-beam evaporation for depositing ohmic contacts to Nb: SrTiO\(_3\) single crystals. Ti thin films are evaporated on the back of Nb: SrTiO\(_3\) to obtain ohmic contact and followed by a thin Au layer to avoid oxidation. Since the work function of Ti is 4.2 eV, it forms a very good large area ohmic contact with Nb: SrTiO\(_3\) whose electron affinity is close to 3.9 eV. Figure 4 (a) shows the AFM image of thin films of Au(15 nm)/ Nb: SrTiO\(_3\) and Fig. 4(b) shows the AFM image of Au(8 nm)/ Cu(15 nm)/ Nb: SrTiO\(_3\) with surface roughness of 0.6 nm and 0.4 nm respectively.

2.3 Film characterization techniques

The as-deposited films are further investigated for their various physical properties. In this section different techniques that are used to investigate thin films are briefly
explained. To investigate the surface roughness of the deposited films as well as of the substrate, we employ the technique of AFM. We then investigate the crystallinity and phase of the film using x-ray diffraction. For further investigation of the crystalline film and the sharpness of the interface at the nanoscale, we have used transmission electron microscopy (TEM).

### 2.3.1 Atomic Force Microscopy

Atomic force microscope (AFM) is the most commonly used technique for studying the roughness and the morphology of the deposited thin films. It is based on sensing the van der Waals forces at the surface of the film [8]. In a basic AFM setup, as the tip scans the surface, the resonance frequency of the cantilever changes due to interactions between the surface atoms and the tip. The cantilever is typically made of Si or Si$_3$N$_4$ with a tip radius of the order of 10 nm. The change in the amplitude of the tip can be detected with a laser light reflected from the tip into an array of photodiodes. An image of the surface is then obtained via a computerized feedback system. The AFM can be operated in a number of modes, depending on the application. But for this work, the measurements are performed using the AFM in the tapping mode, which is more gentle on the sample and the tip than the contact mode. In the tapping mode, the cantilever vibrates with a frequency close to its resonance frequency with a typical amplitude of 100 nm. Due to the forces acting on the cantilever when the tip approaches the surface, the amplitude of the vibration decreases. The height of the cantilever is modified by the system electronics using a piezoelectric actuator, in such a way that the amplitude is constant during the scanning of the surface by the AFM tip. Tracking the cantilever height results in a map of the area that represents the topography of the sample.

### 2.3.2 X-ray diffraction

At the macroscopic scale, the crystal structure is analyzed using X-ray diffraction (XRD). XRD measurements are performed using a Phillips XPert diffractometer, in the group of Prof. B. Noheda at the University of Groningen. The basic principle of x-ray diffraction involves coherent reflection of the incident x-ray beam from a set of parallel lattice planes. In X-ray measurements, one can vary the angles of the sample so as to obtain different kinds of information. A schematic view and the nomenclature of the angles are shown in Fig. 2.5.

1. $\omega$ = Angle between the direction of the incoming beam and sample plane. It can be modified by a rotation of the goniometer around an axis normal to the plane of incidence.
2. Experimental details

**Figure 2.5:** Schematic view of different angles and their relationship to the surface of sample for various XRD measurements.

2. $2\theta = \text{Angle between the incoming x-ray beam and the inspected diffracted direction.}$

3. $\phi = \text{Angle defining the rotation around an axis normal to the sample plane.}$

We explored the diffraction planes that are parallel to the film surface and follow the diffraction condition or Braggs law give as $\sin \theta = \left( \frac{n\lambda}{2d_{hkl}} \right)$ where, $\lambda$ is the wavelength of incoming X-ray ($\lambda = 1.5405 \text{ Å}$ for Cu-K$_\alpha$ radiation and $d_{hkl}$ is the distance between parallel planes in the hkl direction. For our work we have utilized a $\theta$, $2\theta$ scan. In this mode, the X-ray beam is incident at an angle $\theta = \omega$ with respect to the surface plane and the intensity of the reflected beam is measured at an angle $2\theta$ with respect to the incident beam [9]. In analogy with light falling through a grating, the periodicity of the lattice planes and the wave character of the X-ray beam results in a diffraction pattern. The angle $\theta$ at which there will be positive interference between the reflected beams obeys Bragg’s law. Film texture and out-of-plane lattice parameters are evaluated from these scans. (more details in chapter 4)

### 2.3.3 Transmission electron microscopy

For visualization of the crystal structure of epitaxial thin films of LSMO and its interface with Nb:SrTiO$_3$, we use cross-sectional Transmission Electron Microscopy (TEM). The high resolution TEM (HRTEM) images presented in this thesis are measured with a FEI Titan microscope, at the Ludwig-Maximilians Universitat Munchen,
Germany. This system uses a thermally assisted field emission electron gun of energy up to 300 keV (wavelength of $10^{-12}$ m). Using diffraction from such a small wavelength, we can obtain the information down to unit cell size (a resolution of 0.2 nm). It is operated in Bright Field (BF) mode for imaging large areas and HRTEM mode for detailed analysis, such as the thickness of the films, defects, atomic structure, and for investigations of the interfaces. The microscope is fitted with a high angle annular dark field-detector in scanning TEM mode. The cross section specimen is prepared by conventional methods involving cutting, glueing, grinding polishing and ion milling at the Ludwig-Maximilians Universitat Munchen. HRTEM images of LSMO/Nb:SrTiO$_3$ for various thickness of LSMO are presented in chapter 4.

2.3.4 DC magnetization measurements

In order to study the magnetic properties of the films, we have performed magnetization measurements of thin films using a commercially available Quantum Design Magnetic Property Measurement System (MPMS). The MPMS is a system with controlled temperature and magnetic field platform that can measure the magnetic moment of a sample. The sample is moved through a second order gradiometer of superconducting wire that is coupled through a well-shielded superconducting flux transformer to a dc Superconducting Quantum Interference Device, SQUID. The magnitude of the response of the gradiometer is proportional to the magnetic moment of the sample but assumes the sample to be a point dipole moving through the center of the gradiometer. The sensitivity of the system is down to $10^{-7}$ emu with an absolute error of the order of 1% and a relative error of the order of $10^{-5}$. The samples are inserted in the instrument by suspending them with help of a plastic straw (with a low diamagnetic signal) and a thin brass rod. Most of the measurements presented here are done in a field cooled (FC) condition, where the sample is cooled to 10 K in a field greater than the saturation field (typically 500 Oe). In all magnetization measurements on thin films the diamagnetic contribution from the substrate has been subtracted to extract accurate sample moment.

2.4 Device fabrication

Thin films and their heterostructures are fabricated into small devices in order to measure electrical transport in the films, across their interfaces with Nb:SrTiO$_3$, and across their multilayered structures. We have performed in-plane current transport studies using Hall bars, as well as out-of-plane current transport studies using macroscopic current-voltage measurements and ballistic electron emission microscopy (BEEM). A Schottky barrier is usually formed at the interface of a metallic
film with Nb:SnTiO$_3$, but to observe a clear rectifying transport across such an interface it is desirable to have a small area junction with a low leakage at zero bias. To characterize the interfaces using BEEM, the zero bias resistance assumes critical importance, and in our case should be of the order of $10^9$ Ω (discussed in chapter 3). We have also fabricated devices to study spin transport, using magnetic tunnel junctions.

We have employed wet etching techniques to etch out Au, Cu and LSMO bars on Nb:SnTiO$_3$. Magnetic tunnel junctions are fabricated using LSMO ferromagnetic (FM) electrodes and SrTiO$_3$ tunnel barriers. We have etched the multilayers into pillars using argon ion etching. For the fabrication of small spin valve structures, for nanoscale spin transport studies, wet etching as well as dry etching has been used.

2.4.1 Wet etching using UV and deep UV lithography

Using optical lithography, 100 nm thick Au pads of area 220×300 μm$^2$ are evaporated on metallic films as shown in figure 5. These pads are used as top ohmic contacts for the devices. Using another step of optical lithography and wet chemical etching at room temperature, multiple 250×800 μm$^2$ devices are defined. We use Au etchant to etch Au films whereas, for etching Cu and LSMO films, we use aqua regia. Au etchant is a home-made mixture of KI, I$_2$, glycerol and water. The etch reaction occurs as follows:

$$2Au(s) + I_2(aq) \rightarrow 2AuI_2(aq) (2.3)$$

KI is added to increase the solubility of both iodine and gold iodide (AuI) in water allowing a greater concentration of reactant in the solution. For controlled etch rates for ultra thin films, glycerol is added to slow down the etching rate.

Further, Cu and LSMO both dissolve easily in aqua regia which is a solution of one part of HNO$_3$ and 3 parts of HCl as shown in the reaction below:

$$3Cu + 6HCl + 2HNO_3 \rightarrow 3Cu^{2+} + 6Cl^- + 2NO \uparrow + 4H_2O (2.4)$$

2.4.2 Argon ion etching

Dry etching by Ar$^+$ ion milling is used for cases where chemical etching is not possible. We used an Oxford Ionfab 300 ion beam etching system at MESA$^+$ Institute for Nanotechnology at the University of Twente. Argon gas at a process pressure of $3 \times 10^{-4}$ Torr is used. In the ion source, electrons emitted from a thermionic cathode are accelerated by an electric field between the cathode and anode (300 V, 50 mA).
2.4. Device fabrication

Argon molecules are ionized by energized electrons to form gaseous ions which are then extracted from the ion source chamber in the form of a broad beam and accelerated towards the sample (300 V and 1-2 mA). A neutralizer filament emitting electrons is used to neutralize positive ions (124 mA). In principle, ion milling can be used to etch any sample. However, the etch rate depends on the sputtering yield of the material. One of the major problems with ion milling is the re-deposition of milled material, which is avoided by using angled dry etching. Using the above conditions we obtained etch rates of 16 nm/min for Au, 8 nm/min for Co and 5 nm/min for complex oxides studies in this work. Hall bars of LSMO are etched using dry etching and magneto-electrical properties are measured.

2.4.3 Fabrication of Hall bars

To study the electrical transport properties of epitaxial LSMO films of various thickness grown on SrTiO$_3$ using PLD, we patterned the LSMO films into Hall bars. For patterning Hall bars, we used UV lithography and Ar ion etching as explained above. Top ohmic contacts are of Au as in all other devices. The standard Hall bar device design is as shown in Fig. 2.7. Hall bar measurements are carried out in the Quantum Design Physical Property Measuring System (PPMS) at the University of
2. Experimental details

Figure 2.7: Device fabrication steps followed for obtaining a highly rectifying LSMO/Nb:SrTiO$_3$ Schottky interface. (a) - (e) Steps for fabrication of an ohmic contact on LSMO. (f) - (g) The patterning of LSMO bars. (h) An ohmic contact to Nb:SrTiO$_3$ at the bottom of the device.

Twente. We carried out the temperature and magnetic field dependent electrical transport measurements on LSMO films. Magnetic field is applied in a direction in-plane as well as out-of-the-plane to the film.

2.4.4 BEEM device fabrication steps

Figure 2.6 (a)-(g) shows the steps involved in the device fabrication of BEEM studies. It mainly involves two lithography steps and a back contact deposition.
2.4. Device fabrication

Figure 2.8: Multiple devices fabricated for BEEM measurements on Nb: SrTiO₃. First a 100 nm thick Au film is deposited as a top ohmic contact using a lift-off lithography step; second the metal layer forming a Schottky contact with Nb: SrTiO₃ is etched in the shape of rectangular bars and then finally the bottom ohmic contact (Ti/Au) is deposited at the back of Nb: SrTiO₃.

1. Patterning Au bars for top ohmic contact: 100 nm Au pads of area 220×300 µm², as explained in section 2.4.1, have been used as top ohmic contacts or as tunneling contacts for BEEM measurements.

2. Etching the underlying metal bars on Nb: SrTiO₃ so as to define the area of the M-S interface. For this step either wet or dry Ar ion etching has been utilized as explained in section 2.4.1 and 2.4.2 respectively.

3. Ohmic back contact: To form an ohmic back contact (collector contact) the Nb: SrTiO₃ back side is coated with (50 nm)Ti/(50 nm)Au.

Figure 2.8 shows the multiple devices fabricated for BEEM measurements on Nb: SrTiO₃.

2.4.5 Magnetic tunnel junctions

In order to carry out four probe electrical transport measurements of magnetic tunnel junctions (MTJ’s), UV lithography has been used to pattern the samples into micron size tunnel junctions. We have studied various junction areas ranging from 50 to 400 µm². Device fabrication involves four lithography steps as shown in Fig. 2.9 - Fig. 2.10.

1. Etching multilayer structures in shape of bars, (Fig. 2.9 (a-c)).

2. Etching the pillars of tunnel barrier and top ferromagnet on the base ferromagnetic layer, Fig. 2.9 (d-e).
2. Experimental details

Figure 2.9: Top down device fabrication steps followed for all oxide magnetic tunnel junctions of various sizes: (a-c) First, multilayer bars are etched out using Ar ion etching, in order to separate a few bottom LSMO base electrodes on which tunnel junction are etched later on. (d-e) Etched pillars consisting of top ferromagnetic electrode and SrTiO$_3$ on bottom ferromagnetic LSMO bars. (f) Resist is spin coated on the devices before deposition of ohmic contacts to bottom and top ferromagnets.

3. An insulating layer of 200 nm thick AlO$_x$ is deposited using electron beam evaporation, Fig. 2.9 (f) - Fig. 2.10 (a).
2.5 Physical property measuring system

A Quantum Design Physical Property Measurement System (PPMS) was used to measure electrical and transport properties of our thin films. This is an automated low temperature system for the measurement of magneto-electrical properties. The base unit of the PPMS consists of a cryostat with a superconducting magnet coil. For the work presented in this thesis, a PPMS (9 Tesla) system has been used. The measurements are done in the temperature range 10-300 K and in magnetic fields up to 9 T. Magneto-transport data of LSMO films are recorded using ready-made measurement modules. Au bars are used for ohmic contacts on LSMO grown on SrTiO$_3$ substrates. Electrical contacts are made using Al wire bonding onto the chip carrier.

2.6 Ballistic electron emission microscopy (BEEM): Set up

Ballistic electron emission microscopy (BEEM) is a unique technique to study the nanoscale electronic transport across a wide variety of heterostructures. BEEM is a three-terminal configuration of STM that allows the characterization of electron transport through the buried layers and their interfaces at the nanometer scale [43 - 45]. The BEEM system used here is a commercial RHK-UHV 300, Variable Temperature Ultra-High Vacuum Scanning Tunneling Microscope (VT UHV STM). A back
2. Experimental details

Figure 2.11: Principle of the BEEM experiment. (a) Schematics of the BEEM technique. The sample is a thin metal film deposited on a semiconducting substrate. A PtIr STM tip is used to locally inject electrons into the sample by tunneling at a sample tip bias, $V_T$, between the tip and the film. The electrons transmitted perpendicularly through the metal layer are collected as $I_B$ in the semiconductor with a third (rear) electrical contact.

contact to the sample, in addition to the STM configuration (Fig. 2.11), makes collection of the injected electrons possible. These electrons travel through the buried layers and interfaces and thus enable us to investigate their electronic properties. Here, hot electrons with kinetic energies up to several eV are injected from the STM tip (emitter) across a vacuum tunneling gap into a thin metal (base) layer that forms a Schottky contact on a semiconducting (collector) substrate (chapter 3). Injected hot electrons with the appropriate energy and momentum distribution can traverse the base ballistically over the Schottky barrier into the collector. Details of electron transport using BEEM are discussed in chapter 3.

In the main chamber, a pressure of $10^{-10}$ mBar is maintained by a main valve and a loadlock, which is used for loading and unloading the samples. The main chamber has a storage elevator where we can store six holders for BEEM samples or STM tips. The sample stage can be connected to liquid Nitrogen or Helium cryostats in order to measure at low temperatures. Measurements at cryogenic conditions have the advantage of smaller thermal drift and lower Johnson noise. By utilizing both the heating and cooling options, the sample temperature can be maintained at any point
2.6. Ballistic electron emission microscopy (BEEM): Set up

in the range either from 100 K (when a liquid Nitrogen cryostat is installed) or from 25 K (liquid Helium cryostat present) to 1000 K. For the work which is presented in this thesis, we carried out experiments in the temperature range from 120 K to 300 K. Figure 2.12 (a) shows a schematic illustration of the STM unit and (b) shows a schematic picture of the ultrahigh vacuum (UHV)-STM system (UHV 300). (c) shows a picture of the inside of the main chamber with the beetle scan head on top of a sample holder.

The STM unit stands on three legs on the sample stage during measurements. This type of STM unit is called Walker Beetle Design which has been developed by RHK Inc. Each leg consists of a tube type PZT actuator and a sapphire sphere at its end as shown in Fig. 2.12 (c). The tip is also fixed to another tube PZT actuator. The PZT actuators of the legs are used for tip approach and horizontal motion of the STM unit, coarse motion, while the PZT of the tip is used for scanning and fine horizontal motion of the tip. Such a design of the system has several advantages. Any external motors, sliders, and other parts are not needed. Therefore, the
system is compact with a simple control. Furthermore, the maximum movable dis-
tance of the STM unit is not limited by the performance of the PZT actuator but
depends on the configuration of the STM unit and the sample stage. Since the scan-
ing head stands directly on the sample holder, the vibrations and bubbling noises
which transfer through the copper braid will not directly couple into the tunneling
junction. In addition, due to its compact structure and high resonant frequency, the
scan head rejects external mechanical vibrations. Therefore one can easily achieve
high resolution. Finally, the symmetric geometry provides first order thermal drift
compensation in all three axes, eliminating the need for complex compensation soft-
ware.

The BEEM sample holder used for this work is shown in Fig. 2.13. A BEEM
sample is mounted on the BEEM sample holder. Along with the assembled BEEM
holder a copper basket and a sapphire washer are shown. The copper basket is
mounted in the sample holder such that it is electrically isolated from the sample
holder body as shown in the left bottom of Fig. 2.13. The wire attached to the copper
basket is soldered to the inner connection of the available contacts of the sample

2.7 Magnetic tunnel junction measurement set up

To measure the MTJs after their fabrication, they are glued to a chip carrier where connections to the MTJ device are made using wire bonding (Fig. 2.14 b). The chip carrier is loaded on a holder inside a vacuum tube (less than $10^{-5}$ mBar) and is placed in between the poles of an electro-magnet (fields up to $\pm 0.7$ Tesla). The sample is connected to the measurement instruments via a switch box. We apply a bias using Keithley 6517 A. We perform two probe current-voltage (I-V) measurements as well as four probe measurements in our devices. Generally, two probe measurements are performed to check the contacts and LSMO bars. For checking junction resistance, we always perform four probe measurements. Figure 2.14 (a) shows the MTJ device stack and (b) shows picture of a MTJ device after wire bonding. In the four probe measurement geometry, a bias is applied between the lower electrode and junction contact (1-2), and current is measured between the other lower electrode contact and the second junction contact (3-4).

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**Figure 2.14:** (a) MTJ layer stack after device fabrication. (b) Real device image of MTJ device after wire bonding.

holder. This contact is called the BEEM contact as indicated by an arrow labeled BEEM current. This contact is also isolated from the rest of the sample holder and is used to collected the BEEM current. The sample holder has a sample mounted in the copper basket. On top of the sample a sapphire washer is placed to ensure electrical isolation. The Au contact grounds the top metallic surface of the sample such that a tip bias can be applied between the top contact and the STM tip.
2. Experimental details

2.8 Conclusions

All experimental details related to the deposition of epitaxial and non epitaxial multilayers on Nb: SrTiO$_3$/SrTiO$_3$ single crystal are presented. Various characterization techniques used to investigate structural, magnetic and electronic properties of thin films and interfaces are discussed. Device fabrication of Hall bars, BEEM devices and magnetic tunnel junction is discussed in details. A current perpendicular to plane technique to investigate electron transport, at the nanoscale, called ballistic electron emission microscopy, is presented. In the last part of the chapter, the experimental methods used to investigate tunnel spin polarization of LSMO in magnetic tunnel junctions are discussed.

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