Processing of Novel Topological Insulators in Thin Film Form *Rory Barker*



Topological insulators are materials that have only been discovered recently. These materials are electrical insulators with exotic metallic surface states with applications in spintronics and quantum computers. This research focussed on optimising the fabrication process for thin films of a particular topological insulator - Bi_2Se_3 – by using the physical deposition technique called sputtering. The objective of the project was to obtain as smooth a topology as possible by varying deposition temperature on two different substrates. We discovered that deposition at 320°C on Magnesium Oxide (MgO) gave the best results.

Topological insulators are an exciting, recently discovered form of condensed matter. These materials are electrical insulators in the bulk of the material, with exotic metallic energy states across the surface. Growth of thin films of these materials was the topic of my fourth year Materials Science research project. The surface states of these materials show promise in novel applications such as spintronics and robust quantum computing. The reason for producing topological insulators in thin film form is that patterning devices onto a thin film is a fairly practical approach to producing spintronic devices, which could be widely adopted.

Spintronics is an interesting field that potential for developing future has generations of electronic devices. Whereas electronic devices typically rely on the transfer of electronic charge in an electrical current, spintronics would rely on the transfer of the electronic property called *spin* in a 'spin current'. Spin does not refer to electrons literally spinning, but it is an intrinsic property of an electron, like mass or charge. A topological insulator could be used as a spin generator in a spintronic device because electrical transport along the surface states causes electrons to adopt a particular spin orientation that is dependent on the direction of the current.

Topological insulators were not discovered experimentally, but were discovered through theoretical research¹. topological insulators to be The first discovered. known as '2D topological insulators'. exhibited only topological insulator behaviour in very thin 'quantum' wells' where the metallic surface states only existed in one dimension around the edge of the well. More recently, '3D topological insulators' made of the compounds Bi₂Se₃, Bi₂Te₂, and Sb₂Te₃ that exhibit metallic surface states across the surface were predicted to exist in 2009², and shown to exist experimentally in 2010³.

This project focused on growing thin films of the 3D topological insulator Bi₂Se₃ because the large 'energy band gap' in the bulk of the material makes it the most interesting for possible technological applications. With Bi2Se3, it may to be easier to create a material with a high resistivity in the bulk of the material that does not 'short circuit' the surface energy states.

The particular method for growing thin films of Bi_2Se_3 (composed of the elements bismuth and selenium) is a physical deposition method called sputtering. Sputtering works by forming a piece of material, called a target, made up of the elements that make up the desired film. The chamber in which deposition occurs is pumped down to form a vacuum, and then a small amount of argon gas is passed into the deposition chamber. An electric field is used to form argon plasma, and argon ions are accelerated on to the target. As the argon ions bombard the target, atoms of bismuth and selenium are ejected into the plasma. These bismuth and selenium ions can then land on a substrate, and a film can grow. The ratio of bismuth to selenium ions that end up in the film is not necessarily the same as the target, so getting the correct composition of the film takes some trial and error.





The first task of the project was to grow films of Bi₂Se₃ with the right phase and stoichiometry. The equilibrium phase Bi₂Se₃ has hexagonal crystal symmetry and the atomic structure, depicted in Figure 1, needs to be reasonably phase pure. Stoichiometry across the film needs to be well controlled as even other phases with similar compositions to Bi2Se3 are not topological insulators.

Films were initially grown on scrap silicon to find the right stoichiometry. They were then grown on single crystal silicon with hexagonal surface atomic structure in order to try to encourage 'epitaxial growth', where crystals grow layer by layer to obtain a smooth surface morphology. The films were characterised using Xray diffractometry (XRD), and with a scanning electron microscope (SEM). Within the microscope, the ratio of selenium atoms to bismuth atoms on each film was measured using energy dispersive X-ray (EDX) analysis.

The XRD technique used to characterise the phase of bismuth selenide used θ -2 θ geometry. A schematic of x-ray diffraction in this geometry is shown in Figure 2. This technique works by finding the angles of θ through which an x-ray beam can diffracted so that constructive be The diffraction interference can occur. pattern can then be used to figure out the atomic structure of the material.



EDX works in an electron microscope on the principle that the electron beam in the microscope knocks off electrons from the inner electron shells of atoms, so that an electron at a higher energy level can drop down to refill the lower shell and in the process emitting an X-ray with an energy characteristic of that particular atom. This technique, if calibrated correctly can then determine the atomic composition of a sample.



After some experimentation, a target composition was found that gave the right phase of Bi₂Se₃. Figure 3 shows the x-ray diffraction pattern for the first sample found made of the right phase. Once it became apparent that the right phase was formed the effect of temperature on the films was investigated. The films were grown on both MgO and silicon. By varying the temperature, different film morphologies on both MgO and silicon were obtained as shown in Figure 4.



After growing films using the sputtering technique over the temperature

range 280-410°C, it was apparent that films grown on MgO at 320°C have smoother, flatter surfaces than the others, and may prove to be useful for electronic devices with further processing. After characterising the material for the films using both XRD, and EDX, it was clear that the films yielded the right compositions in the right range for most conditions, while the most crystalline films were formed at 320°C.

Another interesting feature that came out of the XRD scans was that the crystals appeared to be orientated in the direction of the c-axis (see Figure 1). This was interesting since films with grains that are aligned could yield smoother morphologies, implying conduction paths along the surface met fewer grain boundaries, which can affect electrical properties of a sample. This needed to be tested by an X-ray 'texturing' technique which would identify the direction in which the crystals were aligned. By rotating the sample through 360 degrees under an X-ray beam, and varying the incident angle of the x-rays, figures could be drawn to identify the direction of the crystal alignments. Figure 5 shows the 'pole figures'.



Figure 5: films grown on silicon (left) reveal that the hexagonal grains are roughly in the same plane, but in random orientations within that plane. Films grown on MgO (right) reveal that the grains are also in the same plane, but the 12-fold symmetry shows that the hexagonal grains must be in two major orientations.

We found that the crystals on MgO were aligned in two main orientations along the c-axis, while the films on silicon were aligned in the c-axis, but with no favoured orientations in the plane of the film. Electrical resistivities of the bulk material were also tested against temperature in a cryostat. The results are shown in Figure 6. These resistivities are relatively low when compared to other published work such as Hong et al.⁵ who grew Bi_2Se_3 nanoribbons with very low charge carrier densities. This

is likely to be a result of defects in the crystals that allow electrons to move freely. The resistivities shown here may be higher than the material resistivity since the test current will have moved through the path of least resistance through the grains, so they do not reveal anything fundamental about the film material. The resistivity can be increased by removing these defects, or by doping the material with the metal antimony. Ultimately, this research was simply a way of learning how best to process this material. While some progress has been made to start making films with the right compositions and smooth microstructures, additional work is needed for further processing of these materials, such as annealing, and doping with antimony. It may even be the case that a different fabrication process is necessary for making topological insulator devices.



References

1. Moore JE, 2010. The birth of topological insulators. Nature, 464(7286), pp. 194-198.

2. Zhang H, Liu C, Xiao-Liang Q, Dai X, Fang Z and Zhang S, 2009. Topological insulators in Bi2Se3, Bi2Te3 and Sb2Te3 with a single Dirac cone on the surface. Nature Physics, 5(6), pp. 438-442.

3. Zhang Y, He K, Chang C, Song C, Wang L, Chen X et al, 2010. Crossover of the three-dimensional topological insulator Bi2Se3 to the two dimensional limit. Nature Physics Letter, 6, pp. 548--588.

4. Semiletov SA and Pinsker ZG, 1955. The electron

diffraction analysis of the system of alloys Bi-Se. Doklady Akademii Nauk SSSR, 100, pp. 1079-1082. 5. Hong SS, Cha JJ, Kong D and Cui Y, 2012. Ultra-low carrier concentration and surface-dominant transport in antimony-doped Bi2Se3 topological insulator nanoribbons. Nature Communications, 3(757).