CHAPTER IV

EXPERIMENTAL TECHNIQUES

A description of various techniques used in the investigations reported in this thesis, is given in brief. The methods of growing single crystals from the melt or vapour are well-known today. The techniques of Multiple Beam Interferometry, Phase Contrast Microscopy and Light-Profile Microscopy are efficient tools for the optical study of crystal surfaces. A detailed account of various types of furnaces and the methods of growing single crystals is given by Lawson and Nielsen[1]. The discussion of the techniques involved in the optical investigation of crystal surfaces is available in the literature[2,3,4]. Hence the details of the theory and construction of the various apparatus, being standard ones, are not discussed.

(A) CRYSTAL GROWING TECHNIQUES:

In the present investigations, single crystals of antimony were grown from the melt as well as from the vapour phase. The modified form of Bridgman's method of growing single crystals from melt was adopted. In Bridgman's method[5] the furnace is kept steady and the charge is moved, while in Chalmers' modified scheme[6] the charge is kept steady and the furnace is moved horizontally along the charge. The method adopted for growing single crystals from vapour phase was the same as that used by Balasubramanian[7].

(1) Preparation of antimony single crystals by Chalmers' method:

The limitation of the Bridgman's technique that it can be used only for the low-melting point metals is overcome by the horizontal furnace moving technique given by Chalmers[6]. The metal is kept in a specially designed graphite boat shown in fig. IV-1. This graphite boat is pointed at one end and the other end is flat. The tip of the boat provides the freezing of the melt at a point because of constriction. In this way a very few seed crystals are formed. The graphite boat is to be kept at the centre of a silica tube, 30" in length and 1" in diameter. The trolley furnace which has to move along the silica tube was prepared according to the standard procedure. The arrangement is shown in fig. IV-2 and a schematic diagram of the same is given in fig. IV-3. X, Y and Z

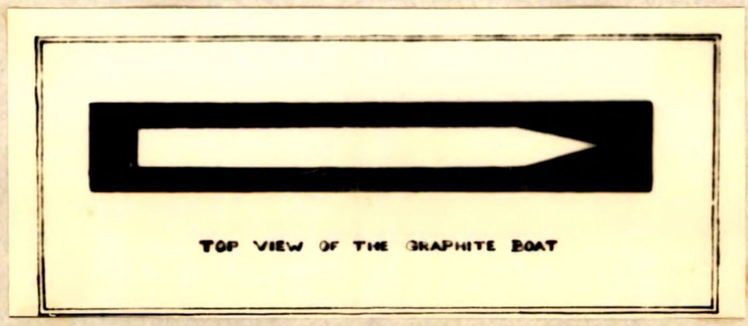

Fig. I

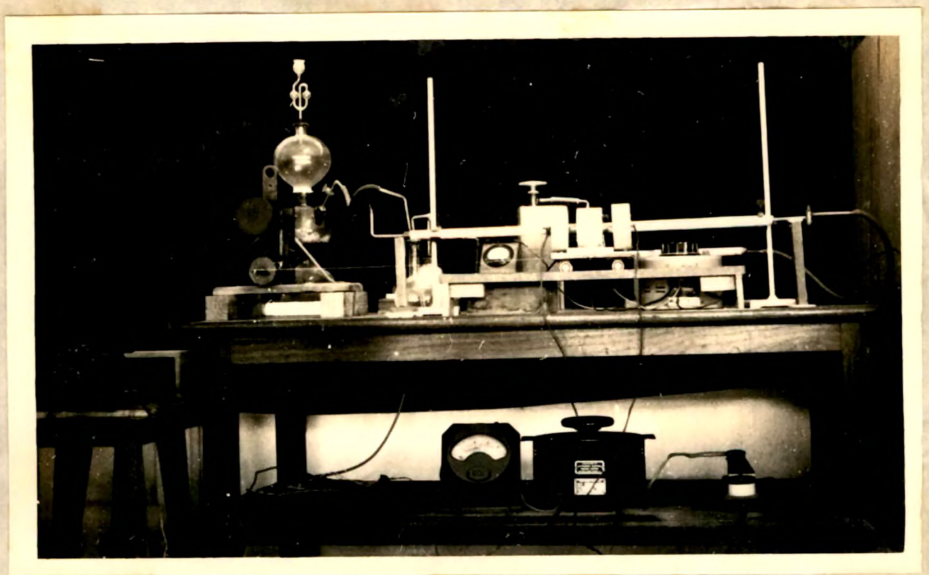

Fig. 2

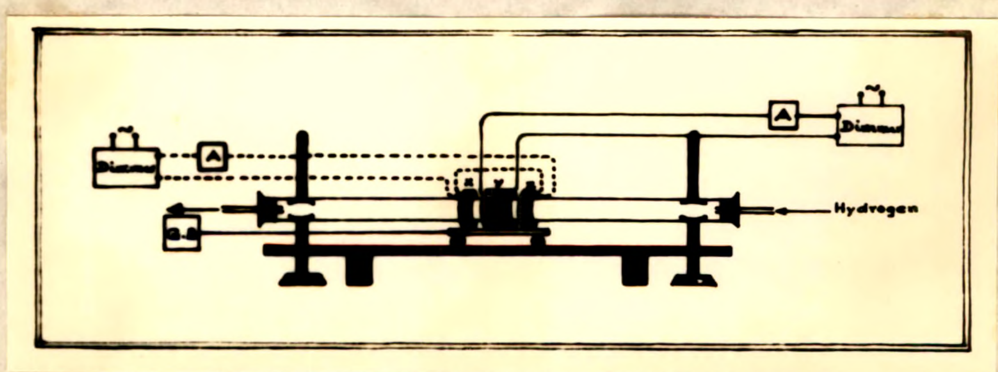

Fig. 3

are three different furnaces where X and Z are connected in series and a separate power supply is used for Y. X and Z are the pre-heating and after-heating furnaces respectively, their temperature being less than the melting point of antimony. The central furnace, Y, is kept at a temperature 50 to 70°C above the melting point of antimony. The temperature gradient can be controlled by changing the temperature of X and Z and also by changing their distances from the central furnace Y. The horizontal movement of the furnace is facilitated by a gear mechanism coupled with a motor. A wide range of rates of motion of the furnace can be obtained by this arrangement. During the growth of the crystals a continuous flow of hydrogen is maintained over the charge. The gas is produced in a Kipp's apparatus and subsequently passed through a column of water to remove the HCl vapour and through a tower containing $CaCl_2$ to absorb the moisture. The gas is then passed over hot copper filings kept in a separate furnace to remove the traces of oxygen present in it.

(2) Preparation of antimony single crystals from vapour phase:

Fig. IV-4 gives an arrangement used for the growth of the crystals from the vapour phase, while fig. IV-5 is a line diagram of the same. The central portion of the fused silica tube, 30" in length and 1.5" in diameter, forms the core of the furnace. The tube is connected to a vacuum system. The other end of the tube can be either closed or connected to a source of dry argon as shown in fig. IV-4. The temperature inside the furnace is measured by means of a thermocouple. A graphite crucible of the size 1" x $\frac{1}{4}$" x $\frac{1}{4}$" with a point bottom is used for growing crystals. This special shape of the crucible occasionally enables one to get the single crystals from melt also. The crucible filled with pure antimony (99.99%) is covered with a graphite plate and is kept at the centre of the furnace. The furnace is continuously evacuated by means of a Cenco Hyvac pump till the pressure is less than 0.1 mm. of Hg. At this stage a stream of argon gas is sent through the furnace to remove the slight traces of air remaining inside it. A steady

Fig. 4

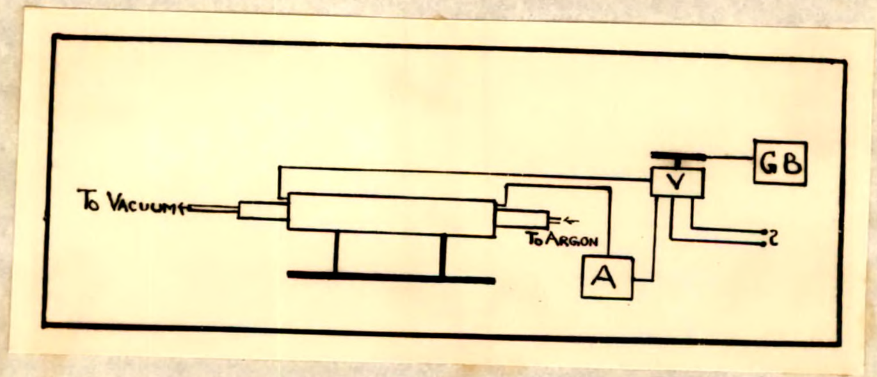

Fig. 5

temperature of about 650° - 700°C of the furnace is attained within 3 hours. The furnace is then cooled down by reducing the current through the coil of the furnace. A steady reduction of the current is made possible by an arrangement shown in fig. IV-4. It consists of a large wheel, 18" in diameter, attached to a variac. This wheel is rotated by means of a gear arrangement coupled with a motor, thereby reducing the current. This mechanism enables a gradual reduction in the temperature of the furnace with a large number of rates.

Generally micro crystals are obtained and they are formed (i) at the end of the graphite crucible near to the pump and (ii) at the cooler end of the silica tube. Occasionally the charge, remaining inside the crucible, solidifies in single crystals. The former two are the results of the vapour phase growth while the last one is the growth from melt which is possible due to the shape of the crucible. The microcrystals have triangular faces with sides 1 to 3 mm. The solid metal which remains inside the crucible is generally polycrystalline one.

(3) Thermal etching equipment:

The furnace used for growing single crystals from the vapour phase has been used for the etching of antimony crystals thermally. The furnace is continuously evacuated through out the process of etching. The air is expelled by flushing dry argon through the furnace before heating it to the required temperature. The cooling of the furnace is done suddenly. Thermal etch patterns are obtained on the surface of the specimen. A very thin amorphous layer of metal is simultaneously observed at the end of the crucible which points towards the pump. It is presumed that the atoms evaporated from the surface are forced to move towards the end of the crucible, due to the pumping, and gathered to form the layer.

(B) TECHNIQUES FOR OPTICAL STUDIES:

Microscopic study of the crystal surfaces has been carried out by means of the Vickers Projection microscope. Phase-contrast observations were made by using the Cooke Phase-contrast unit attached to the Projection microscope. The technique of multiple beam interferometry was used to measure the surface distortion associated with twinning, cleavage steps etc. The optical flats used in these studies were silvered in the Edwards' coating unit. Light Profile Microscopy developed by Tolansky has been used for measuring pit-depths and heights of growth steps. These techniques are well-established ones and their applications are discussed at length in literature[2,3,4]. It is not dealt with in detail here. Attempt is not made to present the theories on which they are based, but a brief discussion is devoted to each of them.

(1) Vickers Projection Microscope:

Vickers Projection Microscope is an inverted metallurgical type microscope which has two different systems for the optical examination of the crystals, one of them being reflection and the other transmission. Since the work presented in this thesis is of metal crystals, only the reflection system was used.

(2) Phase Contrast Microscopy:

The equipment of the Phase Contrast Microscopy can be fitted on the Vickers' Projection Microscope, which is used for the observation of crystal surfaces with high resolution and contrast. The principle of this method is to change the phase variations on the wavefronts leaving the surface into variations in intensity in the plane of the image. Verma[4] has discussed the Phase Contrast equipment which has been used by this author in the course of study.

(3) Multiple Beam Interferometry:

Multiple Beam Interferometry technique is used for the quantitative measurements in the optical study of the crystal surfaces. All the observations were made in the reflected system. Fizeau fringes are obtained by matching the surface under study against a silvered optically flat glass plate. A detailed discussion of this technique is given by Tolansky[2]. The sharpness of the fringe patterns is achieved under the following conditions: (a) the separation between the two surfaces should be of the order of a few wavelengths of light, (b) the surfaces should be highly reflecting with minimum absorption, (c) the optical flat plate should have highly uniform thickness of coating, (d) light should be monochromatic, (e) the beam of light should be parallel and (f) the incidence should be normal.

(4) Light Profile Microscopy:

Light Profile Microscopy is used when the surfaces are very coarse, where the multiple beam interferometry cannot be used. Originally the method requires the projection of an image of the slit on the surface under study. The image of the profile appears as a bright line against a dark background and the magnification is limited to about x400, which makes the surface under study very difficult to observe. Tolansky[3] modified this technique in order to make the study easier. Instead of a slit, a fine wire or a fine scratch on a glass plate is employed, the image of which is projected onto the surface of the specimen by an off-centre illumination. The profile used in the present study was supplied along with the Vickers Projection Microscope. Opaque surfaces can be studied directly, but transparent surfaces have to be silvered for their profile study.

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