PART II

EXPERIMENTAL TECHNIQUES

CHAPTER 2

EXPERIMENTAL TECHNIQUE

The quantitative measurements, in the present investigation, on the growth, the cleavage and the etch patterns produced on the cleavage surface, were made by using multiple beam interferometry, and Light Profile Microscopy. Only short account of these methods will be given here. The optical studies were made either in bright field illumination or phase contrast illumination.

Multiple-Beam-Interferometry:

The first theoretical investigation of multiple beam interference taking place between plane parallel surfaces was made by Airy (1831). The salient points of his work which are of special interest in the present investigation are discussed below. A full account of this technique including the references to original papers is described by Tolansky (1948) and the material is drawn freely from his work. Kuhn (1951) has also reviewed these techniques.

Multiple-reflections taking place between two reflecting surfaces A and B, separated by a medium of refractive index μ and thickness t are shown in fig.Ia . For a beam of light incident at an angle ϕ , let the reflection and transmission coefficients be respectively R and T. Along A there will be a series of reflected beams of intensities R, RT^2, R^3T^2, R^5T^2, etc., and along B with intensities T^2, R^2T^2, R^4T^2, etc. The path difference between any two successive beams along either of the surfaces will be 2ut Cos . The resulting phase lag δ between them will be $(2\pi/\lambda)\,2\mu t$ Cosϕ .

When monochromatic light from an extended source, is directed on to the interferometer, modified transmission Haidenger fringes are formed. If a lens is placed in the path of such a system of parallel rays, Airy summation at the focus of the lens is automatically obtained. The intensity distribution is no longer of Cos^2 type as in the case of two beam fringes but is modified by the multiple beam combination.

The resulting intensity I_t for the transmitted series at any point in the field corresponding to is given by the following relation.:-

$$I_t = \frac{T^2}{(1-R)^2} \quad \frac{1}{1 + \frac{4R}{(1-R)^2} \quad \mathrm{Sin}^2(\delta/2)} \quad \ldots\ldots(1)$$

and for the reflected system

$$I_r = \frac{4R\mathrm{Sin}^2\,(\delta/2)}{(1-R)^2 + \;4R\mathrm{Sin}^2(\delta/2)} \quad \ldots\ldots\ldots\ldots\ldots(2)$$

For a transmitted series I_t becomes maximum, when Sin^2 $(\delta/2) = 0$.

$$(I_t)\mathrm{max} \;=\; \frac{T^2}{(1-R)^2} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots(3)$$

When Sin^2 (/2) = 1, I_t becomes minimum,

$$(I_t)\mathrm{min} \;=\; \frac{T^2}{(1 + R)^2} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots(4)$$

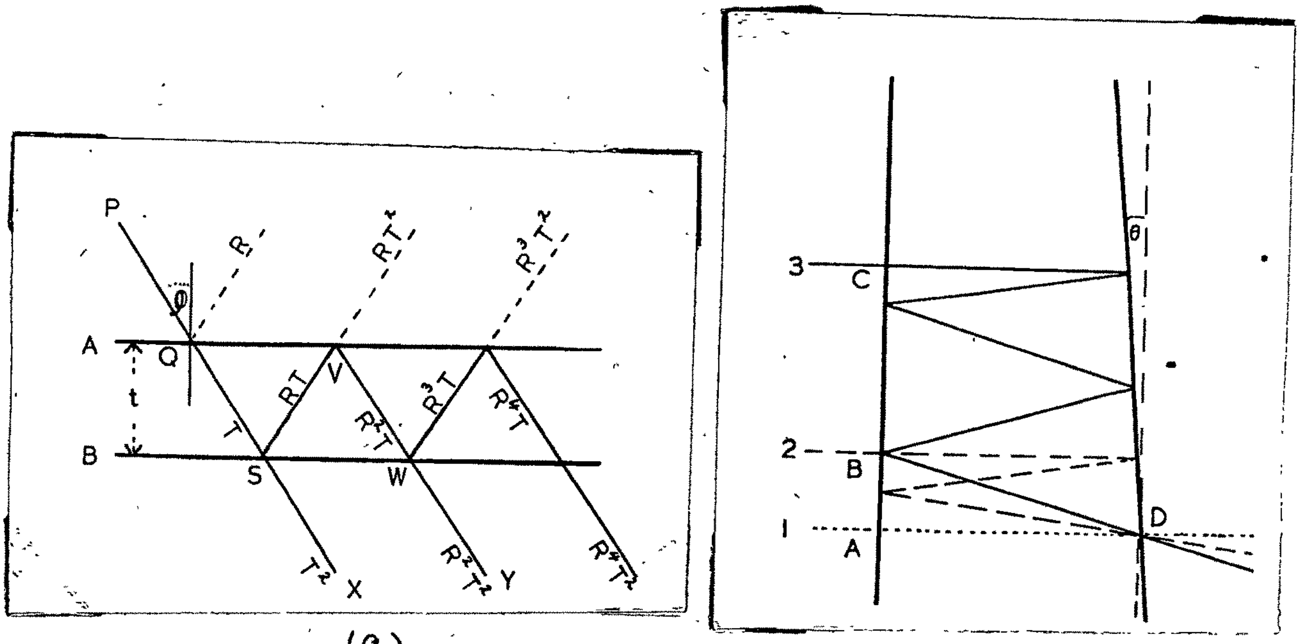
If there is no absorption, $T + R = 1$ and hence the intensity of the fringe maximum in transmission becomes equal to that of the incident light, irrespective of the reflection coefficient R.

If there is absorption A,

$T + R + A = 1$

and hence

$$(I_t)\ \mathrm{max} \;=\; \left[\frac{1 - R - A}{1 - R}\right]^2 \ldots\ldots\ldots\ldots\ldots\ldots(5)$$



(a)

(b)

Fig 1

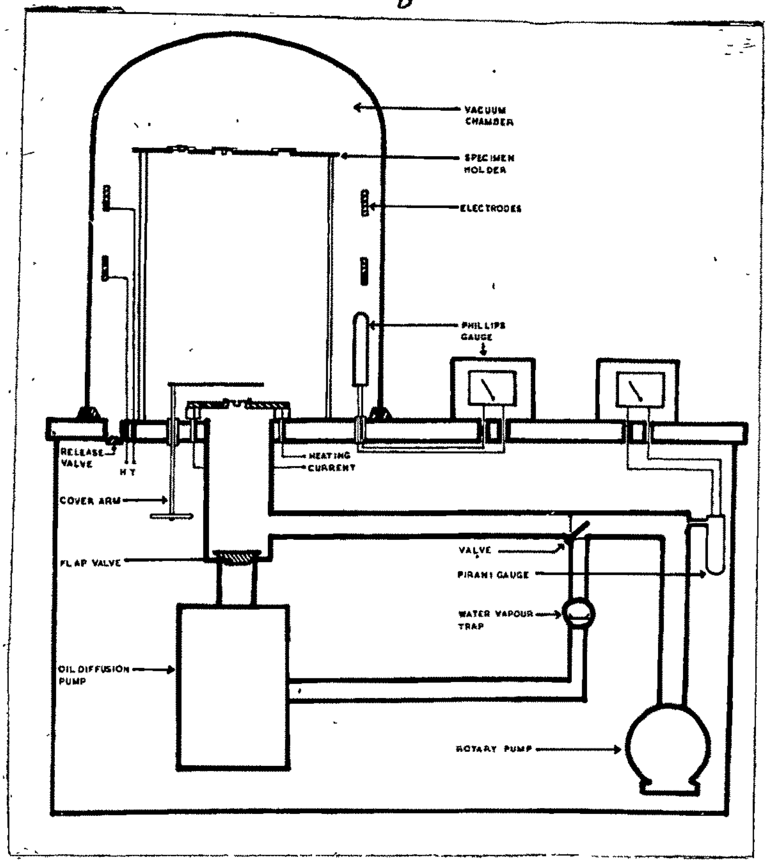


Fig 2a

$$(I_t)\text{min} = \left[\frac{1 - R - A}{1 + R}\right]^2 \quad \ldots\ldots\ldots\ldots\ldots (6)$$

$$\frac{(I_t)\text{max}}{(I_t)\text{min}} = \left[\frac{1 + R}{1 - R}\right]^2 \quad \ldots\ldots\ldots\ldots\ldots (7)$$

The whole fringe shape is thus quite independent of the absorption.

The principal factors affecting the usefulness of multiple-beam fringes are the contrast and the sharpness. The contrast may be defined by term $(I_t)\text{max} - (I_t)\text{min}$ and the sharpness by the reciprocal of the fringe half-width, $W_{\frac{1}{2}}$, which is the width at half the peak intensity.

The theory for the reflected system with films deposited by evaporation has been worked out by Holden (1949) and by Hamy (1906) for chemically deposited films. For this system the value of $(I_t)\text{max}$ decreases while that of $(I_t)\text{min}$ increases with increase in the absorption. The contrast $(I_t\text{max} - I_t\text{min})$, therefore, decreases with increase in the absorption.

The fringe half width, W as a fraction of an order can be shown to be equal to:

$$W \quad \frac{1-R}{\pi\sqrt{R}} \quad \ldots\ldots\ldots\ldots\ldots\ldots(8)$$

The following table gives the approximate value of W for a number of values of R.

TABLE 1

R	0.04	0.7	0.8	0.85	0.9	0.925	0.94
W	1/3	1/9	1/14	1/19	1/30	1/40	1/50

It can be seen from the table that increasing the value of R, decreases the value of W and hence increases the sharpness of the fringes.

The relation between reflectivities, transmission and absorption for silver films is shown in the table 2, below.:

TABLE 2

R %	70	75	80	85	90	94
T %	27	22	16.5	10.5	4.5	0.7
A %	3	3	3.5	4.5	5.5	5.3

This shows that with increase in the reflectivity, the absorption also increases. The increase in the absorption reduces the contrast of the reflected system. In actual practice for the reflected system, it is found that the value of R of about 80 per cent, gives quite sharp fringes with optimum contrast.

Fabry, Perot and Buisson in (1906) produced sharp Fizeau fringes, formed by a thin wedge silvered on both the sides. Tolansky (1948a) was the first who investigated the critical conditions to be fulfilled by a doubly silvered - wedge, so that a close approximation to the Airy Summation can be achieved.

For two inclined surfaces making a small angle, the path difference between successive reflected beams does not remain constant but it increases progressively with increase in the order of reflection. When the path difference becomes $\lambda/2$, the tendency of the beams will be to destroy the condition for the formation of sharp interference fringes. It is shown (Tolansky 1948) that the fringes are formed at the surface and for a wedge angle θ, the path difference between the first and the n^{th} beam, when n is large, to a first approximation, becomes equal to $2nt-4n^{3}0^{2}t$, 't' being the thickness of the wedge at the point of incidence of light. It can, therefore, be seen that the retarding lag behind the arithmetical value (for parallel planes) effectively, becomes equal to $(4/3)n^3\theta^2t$. The retarding lag $(4/3)n^3\theta^2t$ shows that the Airy sum condition will in general be secured only when 't' is very small since for high reflectivities 'n' is very large - of the order of 60, - and θ cannot be reduced indefinitely as its value determines the number of fringes per centimetre across the field of view. Taking possible values of 'n' and θ, it can be shown that the separation between the two surfaces must be of the order of a few wave-lengths of light at the most, otherwise the fringe definition suffers severely.

Apart from the phase retardation condition the linear displacement 'd' of the beams also depends on 't'. The linear displacement of the beams for successive reflection is shown in fig.1b. The variation in 'd' for different values of 'n', 't' and 'θ' is $d = 2n\,(n+1)\,\theta\,t$.

If the values of n and θ are fixed, it is seen that 'd' is proportional to 't'. If 't' is of the order of few wave-lengths of light, it can be shown that for fringes 0.1 mm apart, the linear displacement of the beams is about that of the resolving power of a medium power microscope and hence the beams involved in producing a fringe do not produce any confusion.

Taking account of the phase lag variations due to errors in collimation, linear displacement of beams, etc., the experimental condition for the production of highly sharpened multiple-beam Fizeau fringes can be summarized as follows:-

(1) The surfaces must be coated with highly reflecting films having minimum absorption.

(2) The film must contour the surface exactly and be highly uniform in thickness.

(3) Monochromatic light or at the most a few widely spaced monochromatic wave-lengths should be used.

(4) The interfering surfaces must be separated by at the most a few wave-lengths of light.

(5) A parallel beam (within 1^{o}-3^{o} tolerance) should be used.

Silvering Technique:

For multiple-beam-interferometry evaporated silver films are superior to sputtered silver films. When reflecting film is to be evaporated on to a surface, a condition of cleanliness is of paramount importance. Oil and impurity films reduce the reflecting coefficient somewhat and increase the absorption a great deal, both of which are undesirable for producing sharp fringes. The cleaning agents depend upon the type of the surface about to receive the silver.

Optical flats are washed first with concentrated nitric acid and water and then with soap solution and water and then cleaned with hydrogen peroxide and rubbed gently with dry cotton wool. Such cleaning is continued till on gently breathing over the surface, no breath figures are formed.

In the present work, the use of agents for cleaning of the crystals dependsupon the nature of the crystals and is described in the relevant chapters pertaining to the respective crystal. Final cleaning was done by ionic bombardment in the silvering unit by passing a high tension discharge.

For evaporating silver films, a commercial evaporating unit of the type 12 EA manufactured by Edwards and Co., London, was used. The schematic diagram of this unit is shown in the fig. 2a. It consists of a vacuum chamber in the form of a large pyrex bell jar 35.6 cm. in height and 30.5 cm. in diameter resting in an annular recess upon a gasket of neoprene rubber which rests on a horizontal metal base plate. The chamber is evaculated by a three stage silicon oil diffusion pump backed by a rotary pump. A number of vacuum tight insulated water coöled electrodes pass through the base to which the evaporation filaments (or boat) shown in fig.2b and the electrodes for the high tension discharge are connected. A vacuum tight cone shutter can be swung in and out of position over the filament. The crystal surface or the optical flat on which the film is to be deposited is placed on a tripod, so as to face the filament (or boat). The backing vacuum and the final vacuum can be read directly by the Pirani gauge and the Philips Ionization gauge incorporated in the unit.

The chamber is first evacuated by rotary pump and when a pressure of about .1 mm. of mercury is reached, the high voltage discharge cleaning is started. This is

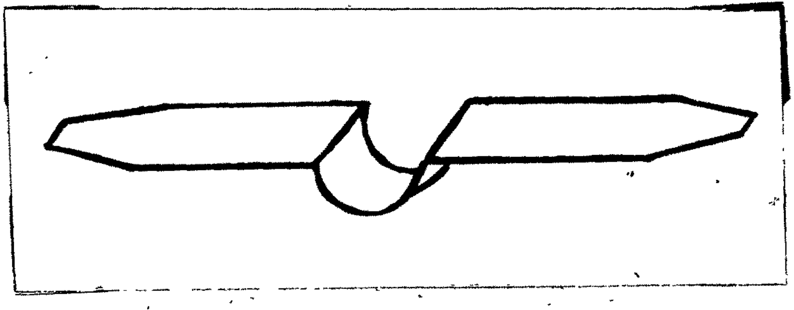


Fig 2b

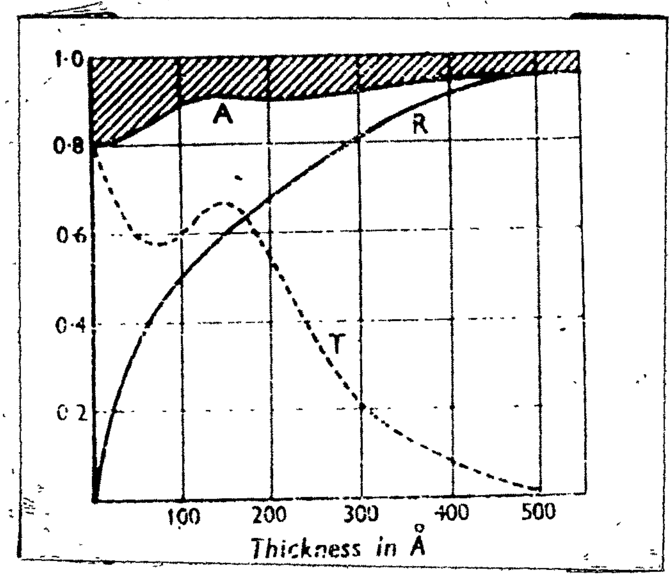


Fig 3

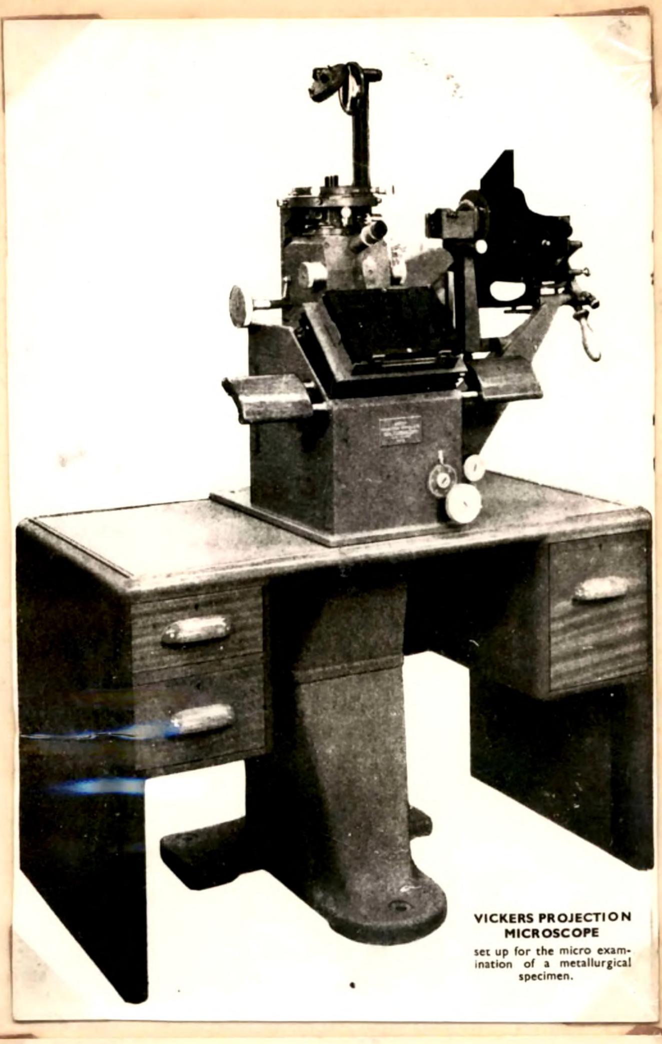
continued until the pressure on the Pirani gauge which shows an increase due to the evolution of absorbed gases after reaching a maximum begins to fall. At this stage the chamber is connected to the diffusion pump. The Philips gauge is switched on, only after the high tension discharge stops because of high vacuum created by the diffusion pump. Silver is generally evaporated when the pressure in the chamber which is indicated by the Philips Ionization gauge, falls to below 10^{-5} cm of mercury. When this pressure is reached, the molybdenum filament (or boat) is heated by a current of about 50 amps. The filament is covered with an adjustable shutter in order to protect the substrate from receiving the vapour of the burnt impurities, Such impurities can have a serious influence in increasing film absorption. Deposition of silver is started, half a minute after the silver starts boiling, which could be seen from outside by removing the shutter from over the filament for the required time.

The thickness of the silver layer and hence the reflectivity is judged either by viewing the filament from the top of the bell jar through the specimen, if the specimen is transparent or, if the filament is calibrated, it can be judged from the time of evaporation.

With pure silver it is possible to get a reflection coefficient of 0.94 in the green and a reasonable transmission of one per cent. Since, now-a-days, powerful sources are available, such reflectivities can be used. The dependence of the reflectivity R, transmission T and the absorption A for different thicknesses of silver films is shown in fig. 3. To obtain high reflectivities, say 90 per cent, the thickness of the silver film should be nearly 500 A^{o}.

Vickers Projection Microscope:

For interferometric as well as microtopographical studies, the Vicker's Projection Microscope (fig. 4) was used. This is of the inverted metallurgical type, the specimen, being placed above the objective. The main collimating system of the microscope is carried on a movable arm to facilitate the change over from reflection system to transmission system. In the present work, the cleavage surfaces of crystals and their etch patterns were conveniently studied with the reflection system. This microscope consists of a universal illuminator consisting of a



VICKERS PROJECTION MICROSCOPE
set up for the micro examination of a metallurgical specimen.

Fig. 4

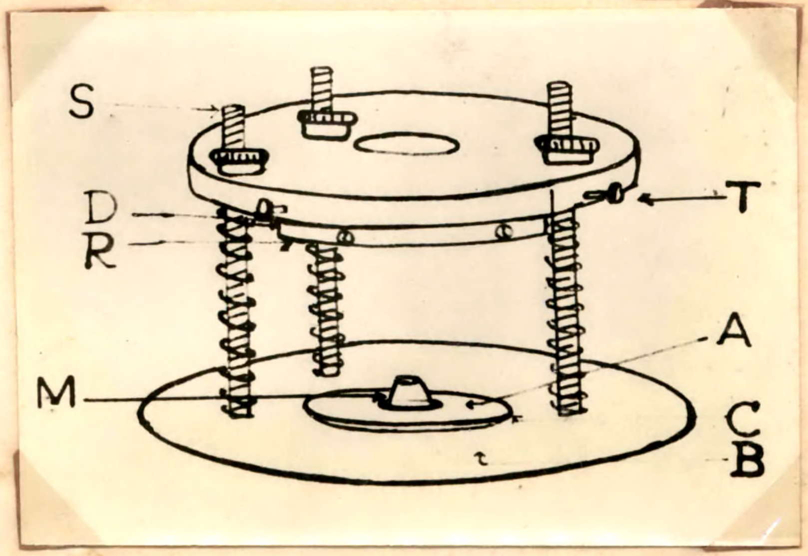


Fig. 5

semi-reflecting plate, iris lens and the objective lens. In reflection the objective thus acts as a condenser, and the convergence of the incident light depends on the relative positions of the back focal plane and the virtual image formed by the field condenser. When these are coincident, a parallel beam falls on the specimen.

The collimating system of the microscope consists of a powerful mercury lamp, a condenser and an aperture controlled by an iris diaphragm, Translational movement of the lamp permits centering of the diaphragm. An image of the source can be formed on the field iris. Movement of the lamp and condenser parallel to the axis of the microscope, makes it possible to vary the convergence of light incident on the specimen.

For visual observations, an eyepiece with a reflector is pushed into the tube below the objective. This completes the normal microscope system. For photomicrography, a projection eye-piece is used and the final image is formed, after reflection in the projection mirror, ~~in~~ in the plane of the screen. Very slight refocussing is necessary when the visual system is changed over to the projection system.

For interferometric work, it is clear from the discussion on multiple-beam-interferometry that for a very small angle between the crystal surface and the optical flat, widely spaced fringes will be obtained. Furthermore, in accordance with the conditions stated for the production of highly sharpened multiple-beam fringes, the separation between the two surfaces has to be reduced to as small a value as possible, normally only 2 or 3 wave-lengths of light. This could be achieved by construct-ing a jig shown in fig. 5, consisting of two circular brass plates of about 1½" diameter, both the brass plates bored in the centre, the diameter of the holes being about 1". In one hole is fitted the glass optical flat in a groove at the base whereas in the other is placed a crystal cemented on a glass plate. These two brass plates are separated by three springs placed at the corners of an equilateral triangle, and can be brought close to one another and inclined to each other at small angles by adjusting the three nuts provided, which press against the three springs. Prior to the arrival of Vickers Projection Microscope, the author was using the metallurgical microscope, with the jig as shown in the figure with the crystal mounted. Since Vickers Projection Microscope is of the inverted type, it is necessary to interchange the positions of the optical flat and the crystal. The same jig was then used with the microscope. Both the flat and the crystal surface were silvered, having the appropriate film thickness, by the silvering technique described above.

Light Profile Microscopy:

The light profile technique of Tolansky (1952) is an improvement and development of Schmaltz (1936) light cut method, then in use for the study of surface profile. In Schmaltz's light cut method, an image of an illuminated slit is projected with a microscope objective at an angle of incidence of 45° on the surface under examination. The image of this surface is viewed with a microscope, the axis of which is at right angles to the first. The profile in depth is converted in appearance into a corrugation in extension. This method gave magnifications in extension upto 400 and in depth up to 650.

Schmaltz light cut technique has many limitations and draw backs such as:

(1) Magnifications are restricted upto × 400.

(2) A special apparatus is required to be prepared with two microscopes at right angles to each other.

(3) Non-specular regions give a false representation.

(4) Surface under examination cannot be seen along with the image of the light cut.

These limitations and draw backs were overcome by Tolansky (1952) in a high resolution light cut and light profile. In devising these techniques, the following important points were taken into consideration:-

(1) Well-defined slit image should be formed.

(2) Single lens should be used for both illumination and viewing.

(3) It is necessary to obtain an off centre pencil illumination in order to secure the condition of

light cut.

(4) Monochromatic radiation should be used to avoid the chromatic difficulties.

(5) Metallurgical microscope should be used.

Any metallurgical microscope by adapting the internal metal tongue reflector, can be readily adapted for high resolution light cut. In this method a slit is made of pieces of razor blade which need not be necessarily narrow and is placed close to the field iris. The author had used an ordinary metallurgical microscope for light profile microscope prior to getting the Vickers Projection Microscope. This was done by using 30° sector reflector. In the Vicker's Projection Microscope the metal tongue internal reflector, which is a metallized sector of about 30° angle, sends an off-centred pencil which is incident on the specimen at an angle of about 40°, when high power lens is used. In this method the objective serves a double purpose of bringing in an off-centred pencil and passing out to the observer E. The experimental arrangement for light profile is shown in figure 6.

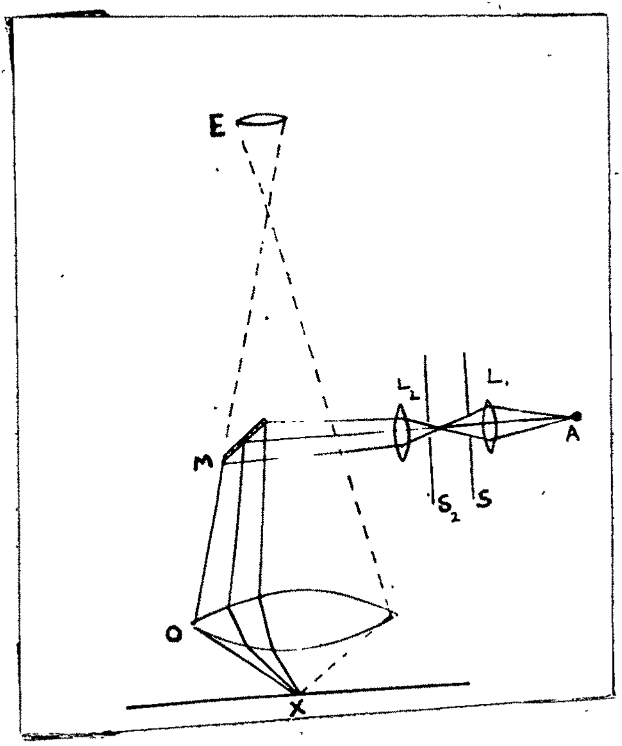

E
L2
L1
A
M
S2
S
O
X

Fig 6

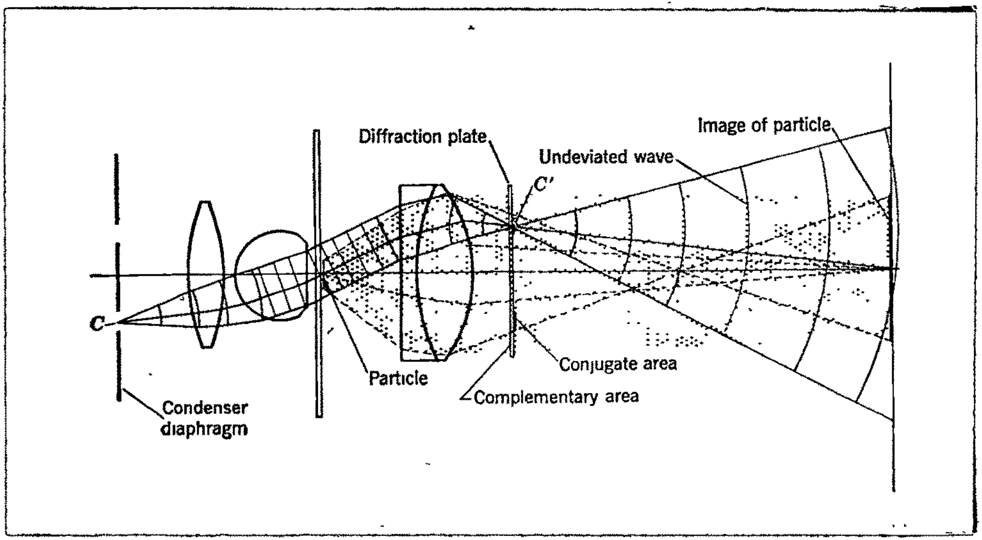

Diffraction plate
Image of particle
Undeviated wave
C'
C
Particle
Conjugate area
Complementary area
Condenser diaphragm

Fig 7a

Light from A which is a monochromatic source passes through the condenser L_1 and the diaphragm S_1. It then passes through S_2 which is a field iris in the universal illuminator. The profile which consists of a mounted thin wire or a scratch on a disc of glass is placed as near to S_2 as possible. The metal tongue reflector M of an illuminator sends an off-centred pencil as shown in the fig. 6.

The procedure of investigating the profile of the surface under examination is very simple. The surface to be studied is placed at X and the microscope is focussed to get a clear image of the surface in the eye-piece E. The profile is now inserted near to S_2, and is moved forward and backwards till the sharp image of the profile is formed on the surface. The profile appears as a dark sharp line, corrugated in extension for the surface features in depth and height. From the shift in the profile, the actual depth or height can be easily calculated.

If 'm' is the linear magnification of the microscope in extension, i is the effective angle of incidence of the pencil and n is the refractive index of the medium between the work piece and the objective, the magnification of the profile will be (2m/n) tan i. In this expression (2/n) tan i is a constant quantity for a particular objective, which can be readily evaluated by interferometric calibration. The values of the constant (2/n) tan i for different objectives used with the microscope in this work are as follows.:-

Objective	Oil Immersion	4 mm.	8 mm.
Constant 2/n tan i	1	1.38	0.56

Knowing the value of the constant 2/n tan i and the linear magnification 'm' of the microscope in extension, the magnification of the profile can easily be calculated.

Magnification up to X 2000 both in extension and in depth can be used and a full aperture is available for the formation of the image. This technique can well be used for both transparent and opaque substances. In order to obtain the requisite contrast at such high resolution and magnification it is necessary to evaporate silver on the surfaces of all the substances to be studied by this technique.

Phase Contrast Microscopy:

The function of an ordinary microscope is the formation of an image in terms of brightness of colour contrast which can be converted into an image that can be observed. The change in the brightness of a light wave is caused by the change in amplitude as a result of absorption of the light and the change in the colour is produced as a result of selective absorption. This means that the changes produced by an object in the amplitude and the wavelength of a light wave can be easily detected by an ordinary microscope.

The optical path differences introduced by the microscopic specimen, from the difference in the refractive index or the thickness or a combination of both, cannot be detected by an ordinary microscope. The possibility of revealing those parts of the microscope specimen having different refractive indices, compared to their surroundings, as a change in the intensity of the image was first investigated by Zernike (1934). The method was later developed by Jupnick, Osterberg and Pride (1946); and

Taylor (1949) for use with specularly reflected objects. A comprehensive review of the subject is given by Bannet, Osterberg, Jupnick and Richards (1951) in their book " "Phase Microscopy". The detailed mathematical theory based on the use of vector diagrams has been published by Barer (1952). A good account is given by Verma (1953).

The function of an ordinary microscope and the alternations required to be made to convert it into a phase microscope can be easily marked in fig. 7a, in which C is a condenser diaphragm. Let us consider the formation of the image of a single transparent particle which is placed at the position shown in the figure. The rays from C are rendered parallel before passing through the specimen. According to the theory of the microscope developed by Abbe (1873), every point in the specimen is treated as a diffracting object. Consequently the incident wave does not pass without interruption through object plane but is diffracted. As shown in the fig. A portion of the diffracted wave, which is shown by solid lines, continues on its original course, undeviated, while the remaining portion shown by the dotted lines gets deviated. The undeviated waves are called S (Surround) waves while the deviated wave is called the D Wave. The S waves from an image of C at C', the back focal planex of the objective and after passing through C', diverge and spread uniformly over the image plane. The D waves spread out over the back focal plane and are focussed by the objective upon the neighbourhood of the geometrical image of the particle. The interaction of the D and S waves in the image plane forms an image.

If the particle is transparent and introduces a small path difference, it can be shown as in figure 7b, that the D wave lags $\lambda/4$ wavelength behind the S wave, when the optical path of the particle exceeds that of the surround. Now D + S = P, the particle wave. The amplitude and the phase of the light that enters the image of the particle, is the amplitude and phase of the P wave and the amplitude and the phase of the light that enters the image of the surround is that of the S wave. Because the light transmission of the particle and the surround is the same, the image of the particle will show no contrast as the amplitude of the P and the S wave are equal.

In the phase contrast microscope, a diffraction

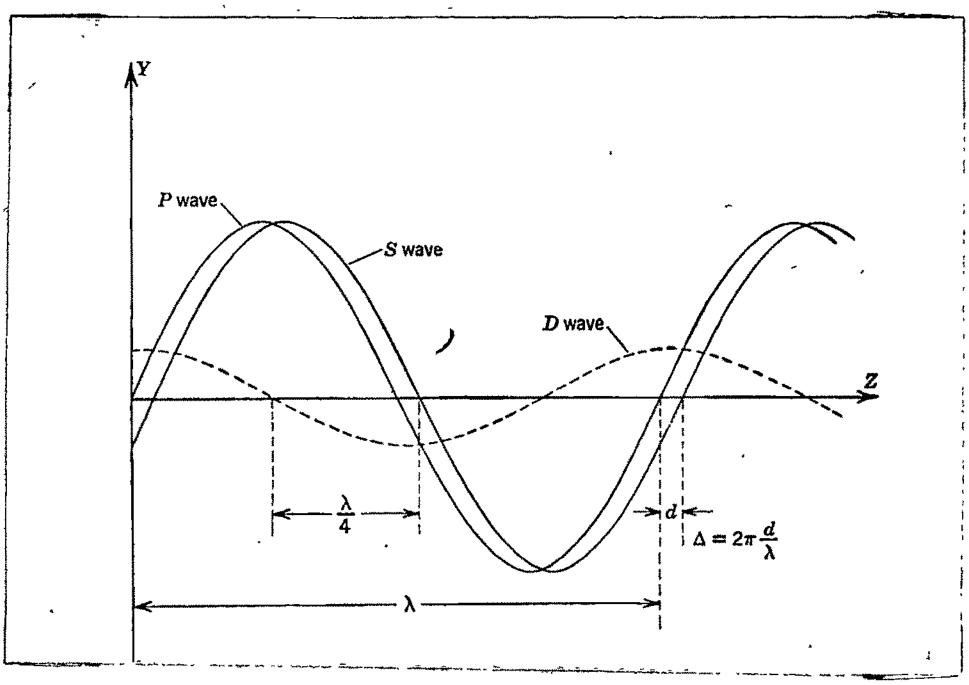

Y
P wave
S wave
D wave
Z
$\frac{\lambda}{4}$
d
$\Delta = 2\pi \frac{d}{\lambda}$
λ

Fig 7b

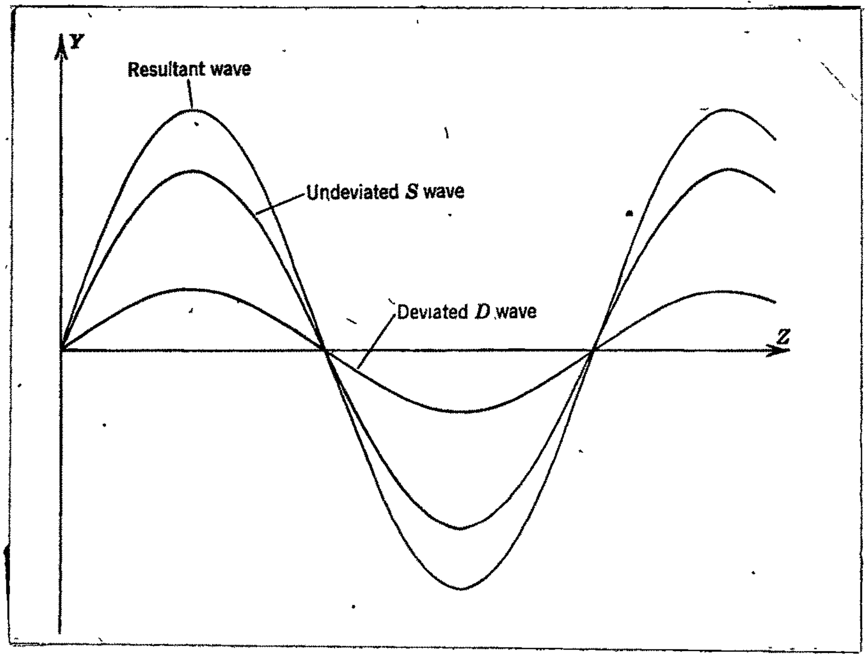

Y
Resultant wave
Undeviated S wave
Deviated D wave
Z

Fig 7c

plate is introduced at C', the back focal plane of the objective as shown in the figure. On the diffraction plate the conjugate area through which the undeviated waves pass coincides with the image of the condenser diaphragm. The conjugate area being small compared with the area of the diffraction plate, the deviated waves pass through the complimentary area, i.e. through that area of the plate which is unoccupied by the image of the opening of the condenser diaphragm.

Now by depositing a suitable coating of reflecting material, such as magnisium fluoride, either on conjugate or complementary area of the diffraction plate, the S wave or the P wave is artificially retarded by $\lambda/4$ wavelength. Figure 7c shows the effect of such a coating over the conjugate area. The undeviated wave S is now in phase with the deviated wave D and therefore the amplitude of the wave $P = D + S$ becomes maximum. Therefore, the particle appears brighter than the surrounding area and this is known as a negative phase contrast.

If on the other hand similar coating is deposited on the complimentary area, the particle will appear darker

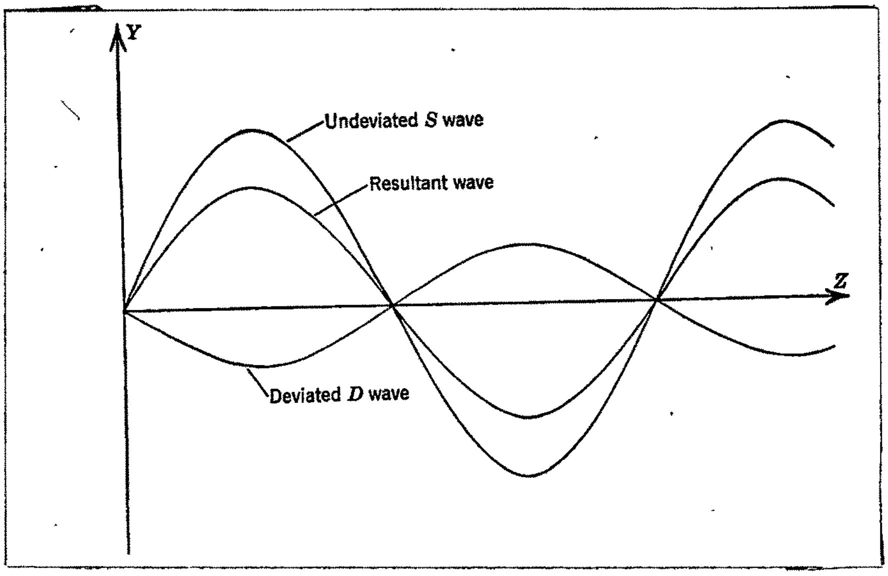

Y
Undeviated S wave
Resultant wave
Z
Deviated D wave

Fig 7d

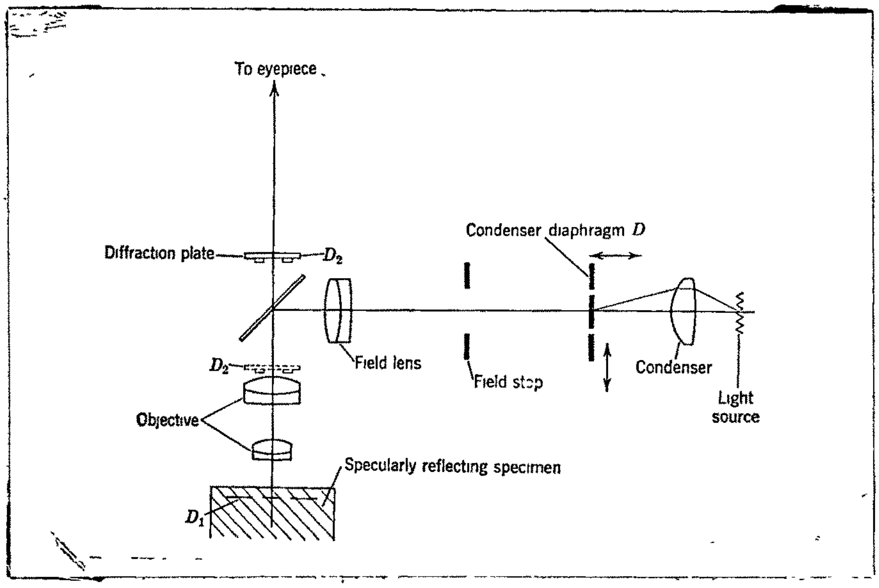

To eyepiece
Condenser diaphragm D
Diffraction plate
D_2
Field lens
Field stop
Condenser
Light source
D_2
Objective
Specularly reflecting specimen
D_1

Fig 7e

than the surrounding area, and this then gives a positive phase contrast fig.7d.

For obtaining optimum contrast in the image the conjugate and the complimentary areas of the diffraction plate are further coated with absorbing materials to equalize the amplitudes of the undeviated waves.

A phase contrast microscope, which was used by the author and found generally satisfactory was the Cooke, Troughton and Simms phase contrast equipment for incident illumination. This equipment was used when fitted on the Vicker's Projection Microscope.

A diagrammatic scheme of the arrangement is shown in figure 7e. An annular diaphragm D serves as the entrance pupil of the optical system which consists of a field lens, the microscope objective, and the reflecting surface of the specimen. The light source is imaged on the diaphragm D by the condenser lens. The field lens and the objective form an image D_1 of the field stop on the specularly reflecting surface of the object specimen. The light is reflected from the surface of the specimen and is then passed through the objective again to form a real image D_2 of the diaphragm D. Image D_2 becomes the exit pupil and therefore the location of the phase plate.

In this equipment, the illuminator tube carries the annulus which, in this case, is the same for all powers of objectives. The annulus is readily moved in and out of action and is provided with a centring adjustment. The illuminator tube also contains a condensing lens and is fitted with a variable power adjustment whereby the annulus may be made to coincide with the phase plates. The annulus is centred on to the phase plates with the aid of an auxillary microscope.

The reflector body consists of objective, adjustable illuminator plate and a slide enabling four systems of illumination to be made available, viz., positive and negative phase contrast, dark ground illumination and normal incident illumination. The slide is located between the illuminator plate and the eye-piece since in this position both the amount of stray light and losses of illumination are minimized.

The unit supporting the reflector body and illuminator tube is attached to the instrument by a bayonet joint and is interchangeable with the universal illuminator

The specimen is supported by a superstage which is clamped firmly to the main stage of the instrument.

Thus this equipment provides the use of a positive or negative phase contrast.

While using phase microscope, it is necessary that the surface of the specimen must be adjusted perpendicular to the optic axis of the microscope, in order to centre the image on the optic axis. An auxillary microscope is used in the place of the eyepiece enabling the examination of the back focal plane of the objective so that the image of the condenser annulus and the phase plate may coincide. The ring form of the condenser annulus together with an annular phase plate has a considerable advantage. Since this form has complete axial symmetry, it introduces practically no undesirable lack of symmetry in the image and the resolution is improved and depth of focus diminished due to the use of large objective aperture by direct light. Furthermore, less diffracted light is wasted owing to overlapping between the higher order spectra and the retarding annulus of the phase plate.