

## The Christiansen experiment with birefringent powders

SIR C V RAMAN and M'R BHAT

(Memoir No. 68 of the Raman Research Institute, Bangalore)

Received February 18, 1955

### 1. Introduction

In the well-known experiment due to Christiansen which has been applied in the construction of monochromatic light-filters, an isotropic transparent solid, e.g., optical glass, is powdered and placed inside a flat-sided glass cell and the latter is then filled up by a liquid whose refractive index is adjusted to equality with that of the solid for some particular wavelength in the spectrum. Christiansen himself tried using powdered quartz in the experiment, and found that it did not prove a success. The light entered the cell and was diffused by the powder-liquid mixture, but the source of light could not itself be seen through the mixture. Considered from the standpoint of geometrical optics, this result is not surprising, since the refractive index of the particles of a birefringent powder would depend on their orientation within the cell and hence would vary from particle to particle.

However, geometrical optics does not correctly describe what is actually observed in the Christiansen experiment even with optically isotropic powders,<sup>1,2</sup> and hence there is no reason to believe that it would be any more successful in the case of birefringent powders. In another paper<sup>3</sup> appearing in the present issue of these *Proceedings*, a general theory has been developed which indicates that chromatic effects analogous to those observed with isotropic powders should also be capable of appearing with birefringent powders in appropriate circumstances. The manner in which the birefringence would modify the observed effects has also been discussed in that paper.

It is proposed in what follows to place on record some observations which we have made broadly confirming the indications of the general theory. It has been found that provided the birefringence is fairly small and the material is in a finely subdivided state, it is possible to obtain a true transmission exhibiting brilliant colours. It is also found that the light so transmitted is not greatly inferior in the degree of its monochromatism to that observed with isotropic material in similar circumstances. Another noteworthy feature is that if the light incident on the cell is plane-polarised, the light transmitted by it is also completely plane-polarised. This might seem paradoxical when it is recalled that the light has had to pass through considerable thicknesses of a birefringent material; nevertheless the

observation is in strict accord with the theory. On the other hand, the diffusion halo appearing in directions surrounding the regularly transmitted light exhibits imperfect polarisation to an extent depending on the fineness of the subdivision of the material. The colours exhibited by the diffusion halo are also found to be markedly different for the components of the light vibration respectively parallel and perpendicular to that in the incident light.

## 2. Some general observations

Besides the factors which determine the transmission coefficient with isotropic powders, an additional factor appears in the present case, namely the magnitude of the birefringence of the material. The importance of this relatively to the differences in the refractive indices of the solid and liquid varies with the proportion of the volumes occupied respectively by the powder and the liquid in the cell. By varying this proportion, we may pass from one extreme case of a polycrystalline aggregate in which the liquid serves merely to secure optical continuity between the particles of the powder to the other extreme case in which the liquid occupies the whole volume except for the particles of solid held in suspension. The most interesting cases are however those in which the two components are present in nearly the same proportions. To observe chromatic effects in such cases, it is necessary to use material which is not too highly birefringent and which is in a fine state of subdivision. The latter condition is most conveniently secured by using a substance which is commercially obtainable in the state of a fine powder and hence does not need any further preparation. We shall content ourselves here by mentioning three such substances which we have found to work very well in the experiment.

Barium sulphate can be used with success in a cell from five to ten millimetres thick with carbon disulphide as the liquid filling it up. The addition of a few drops of benzene shifts the transmission from the yellow towards the violet end of the spectrum. Precipitated calcium sulphate in the form of gypsum also gives good results, the appropriate liquid in this case being monochlorobenzene; a drop or two of carbon disulphide shifts the transmission towards the red, while the addition of a few drops of benzene shifts it towards the violet. Magnesium fluoride also serves admirably; the appropriate liquid to use in this case is acetone, the addition to which of benzene causes the transmission to appear first at the violet end of the spectrum and shifts it step by step toward the red.

## 3. Polarisation phenomena

In all the three cases mentioned, beautiful chromatic haloes are observed surrounding the direction of the transmitted light. The colour of the halo varies

with the direction of observation and also alters when the spectral region of transmission is shifted. Viewing a bright and well-defined light source through the cell held before the eye of the observer, with one polaroid inserted between the source and the cell and a second polaroid between the cell and the observer's eye, striking polarisation phenomena may be observed. In all cases, the image of the light source is itself completely extinguished when the two polaroids are crossed. But the diffusion halo seen surrounding the light-source shows imperfect polarisation. The magnitude of this imperfection differs very much in the case of the three substances mentioned above. With magnesium fluoride the halo disappears almost completely when the polaroids are crossed. With barium sulphate its extinction is less complete, and the diffusion halo remains observable in directions adjacent to the source and gives indications of a bright cross with its arms bisecting the angle between the vibration directions of polariser and analyser. The extinction of the diffused light is least perfect in the case of calcium sulphate and the halo remains observable over the whole area of the field even with the polaroids crossed.

#### 4. Observations with powdered quartz

The following method was adopted to prepare quartz in a state of fine subdivision but uncontaminated by extraneous material. A transparent piece of crystalline quartz was heated and dropped into cold distilled water. The fragments into which it broke as the result of this treatment were heated in a silica dish and then again dropped into cold distilled water. Repetition of this procedure reduced the substance to a state of powder and the material thus obtained was then ground up very fine between two quartz crystals with flat faces and finally separated into four grades by elutriation in distilled water. The two finest grades thus obtained were those which remained in suspension in a tall beaker of distilled water for ten and twenty minutes respectively.

The optical effects exhibited by the four grades of quartz when placed in a cell and filled up with a mixture of benzyl alcohol and carbon disulphide were found to be very different. The first or roughest grade gave only a diffusion halo without any regular transmission even with a cell only one millimetre thick. With the second grade of powder and the same thickness of cell, a very weak transmission can be glimpsed, the bright diffusion halo overlying it making the observation rather difficult. On the other hand, the two finer grades when allowed to settle down in a cell two millimetres thick show brilliantly coloured transmitted images of the source. With a cell only one millimetre thick, the transmission is even more brilliant, but its colour is then less saturated.

In all cases, if a transmission is obtained at all, it is completely extinguished when the cell is placed between two crossed polaroids. On the other hand, the appearance of the diffusion halo as well as its state of polarisation shows

remarkable variations with the grade of powder and the thickness of cell employed. With the coarser grades of material, the halo is found to be completely depolarised. On the other hand, with the finer grades the halo shows a very marked degree of polarisation. Not merely the brightness of the halo but also the distribution of colour in it is strikingly different when the polaroids are respectively parallel and crossed. This difference is best described by the statement that when the polaroids are crossed, the halo exhibits colours similar to those of the transmitted light (which is extinguished in the same circumstances); *per contra*, with the polaroids parallel, the colour of the halo is complementary to that of the transmitted light. These changes were most striking when observed with the cells of smaller thickness. For, with such cells, the colour of the halo over its whole area is markedly different from that of the transmitted light and exhibits its complementary character most clearly. The changes produced by the rotation of the analyser are therefore particularly striking.

### 5. Observations with dilute suspensions

With the two finer grades of quartz powder, it is possible to use much thicker cells with success for observing the transmitted light, if they are filled with an excess of liquid in which the powder is held as a dilute suspension. By varying the thickness of the cell and the quantity of material held suspended in it, one can either increase or decrease the saturation of the colours observed in the transmitted light and in the diffusion halo. It is worthy of note that in such cases the diffusion halo exhibits a colour complementary to that of the transmitted light even in directions adjacent to the latter. This indeed is what optical theory indicates should be the case for a dilute suspension. On the other hand, multiple scattering comes into play in the case of dense aggregates, and the colour of the halo is in consequence almost indistinguishable from that of the transmitted light in closely adjoining directions and only further out changes by insensible gradations to the complementary tint.

It appears worthwhile also to record some observations made with strongly birefringent powders, calcium carbonate in the form of precipitated chalk being a typical example. It is not possible to obtain any transmission with this material in the usual form of the Christiansen experiment. But interesting effects may be observed even with cells of considerable thickness if they are filled with carbon disulphide into which a little precipitated chalk is put in and stirred so that the liquid appears as a milky-white suspension. A bright source of light can be seen through such a suspension and exhibits a deep red colour, as indeed it should according to the theory. Here again the transmitted light is completely extinguished if the cell is placed between crossed polaroids. On the other hand, the light diffused by the cell is depolarised, but if the suspension is very dilute, the cell exhibits a bright cross between crossed polaroids. If benzene is added to the

suspension of chalk in carbon disulphide, thereby bringing the index of the liquid mixture nearer to the mean index of calcium carbonate, the colour of the transmitted light is shifted towards shorter wavelengths in the spectrum.

## 6. Description of the figures in plate I

By way of illustration of the foregoing observations, some photographs have been reproduced in plate I accompanying this paper. The following remarks are explanatory notes on the same.

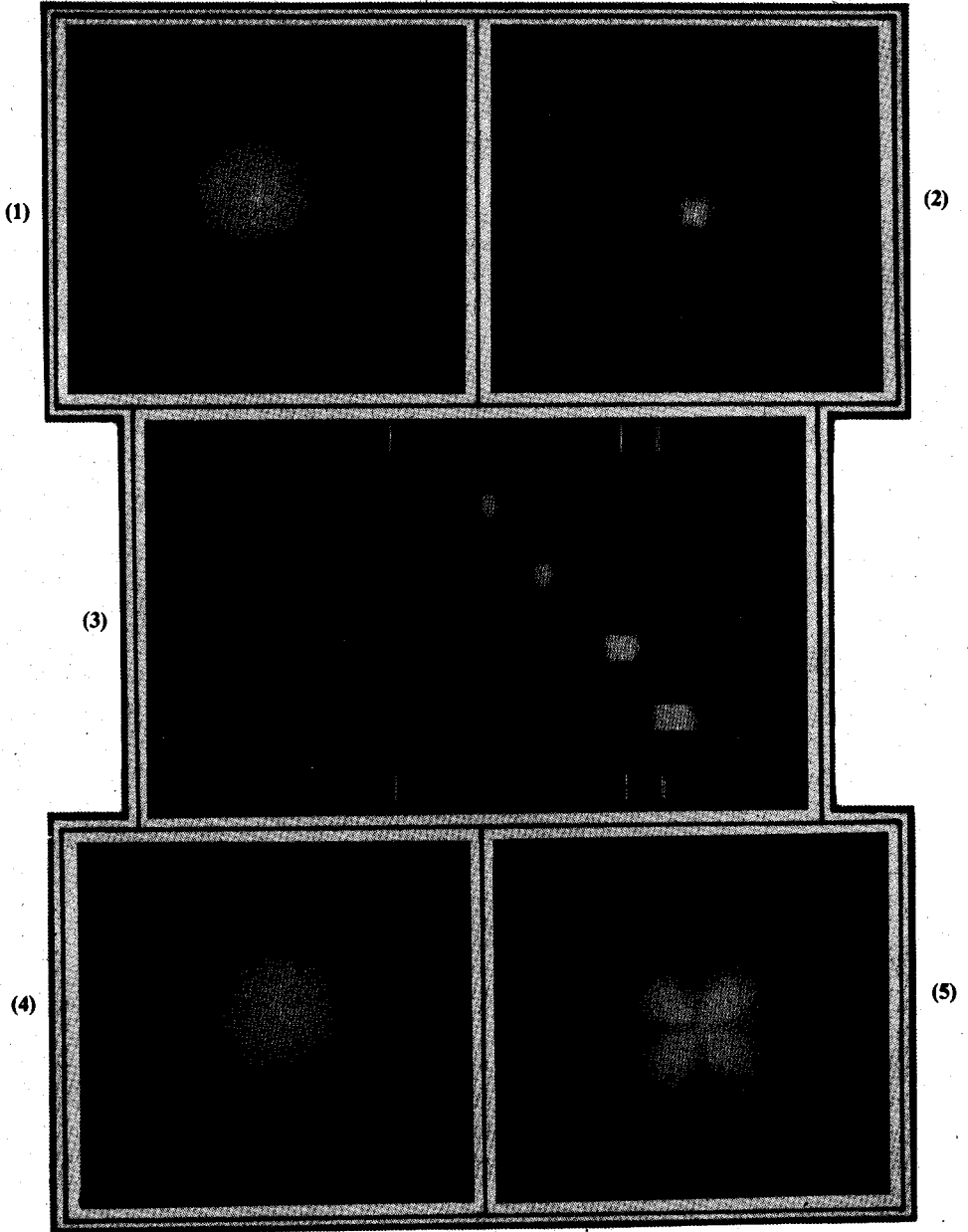
Figure 1 shows the appearance of a small brilliant light-source as viewed through a cell five millimetres thick containing barium sulphate in a finely subdivided state. This had settled down at the bottom of a mixture of carbon disulphide and benzene so adjusted as to transmit the green part of the spectrum. The picture clearly shows the transmitted light and the diffusion halo surrounding it.

Figure 2 shows the same experiment with the cell placed between crossed polaroids and photographed with a much longer exposure. The source itself is extinguished in these circumstances. The halo also disappears except at its brightest part near the centre which exhibits the bright cross whose arms bisect the angle between the vibration directions of the polariser and analyser.

Figure 3 reproduces the spectra of the light transmitted by the cell containing barium sulphate in the same circumstances. The colour of the transmitted light was shifted in steps from orange-yellow to blue by the successive additions of benzene to carbon disulphide. The lines of the mercury arc spectrum are reproduced to indicate the positions of the transmission band.

Figure 4 is a photograph of a small bright source of light seen through a cell two millimetres thick containing the finest grade of quartz powder which had settled down at the bottom of the cell. Benzyl alcohol to which a few drops of carbon disulphide had been added was the liquid used and the mixture transmitted the yellowish-green part of the spectrum. The photograph itself was rather overexposed with the result that the transmission and the halo are not seen clearly distinguished from one another. The streaky nature of the halo is clearly shown.

Figure 5 is a photograph of a bright source of light seen through a thin film of nitrobenzene containing fine particles of lithium carbonate in suspension and held between crossed polaroids. The dark arms of the cross seen in the diffusion-halo are parallel to the vibration directions of the polariser and analyser respectively.



Figures 1-5. Christiansen experiment with birefringent powders.

Plate I

## 7. Summary

Optical effects analogous to those exhibited by isotropic materials in a Christiansen cell are also observable with birefringent materials in a fine state of subdivision. While the transmitted light is fully polarised, the diffusion halo is depolarised in part and exhibits colours between parallel and crossed polaroids which are complementary to each other.

## References

1. C V Raman and M R Bhat, *Curr. Sci.*, 1953, **22**, 31.
2. M R Bhat, *Proc. Indian Acad. Sci.*, 1953, **A38**, 67.
3. C V Raman and K S Viswanathan, *Proc. Indian Acad. Sci.*, 1955, **A41**, 55.