Proc. Indian Acad. Sci. A30 211-215 (1949)

The Christiansen experiment with spherical particles

C V RAMAN

and

S RAMASESHAN

(From the Raman Research Institute and the Department of Physics, Indian Institute of Science, Bangalore)

Received November 5, 1949

1. Introduction

During a visit by the senior author to the works of Messrs. Chance Brothers at Birmingham in May 1948, he noticed in their showroom an exhibit of glass in the form of tiny spherules about a millimetre in diameter. A sample of the material very kindly presented by the firm was brought back to India, and the studies now reported were made with it.

On placing a quantity of the substance in a transparent cell and holding it up against a bright source, it is found that even a moderate thickness of it suffices completely to cut off the light and make the cell opaque. When, however, the empty spaces between the spherules of glass are filled by a mixture of carbon disulphide and acetone of suitably adjusted refractive index, the Christiansen effect is observed. In other words, the cell becomes transparent to a restricted region of the spectrum, while the rest of the light appears as a coloured halo surrounding the source when viewed through the cell. The explanation of these facts is obviously the same as in the usual form of the Christiansen experiment. Nevertheless, the phenomena observed are distinctly more spectacular with the spherules than with a pack of irregular fragments of glass. The spherical shape of the particles influences the propagation of light through the cell in a characteristic fashion and results in some readily observable and rather striking consequences. Then again, the uniformity of size of the particles results in a high degree of regularity in their disposition within the cell, and this becomes conspicuously evident in the experiment.

Though the material as received shows the effects in question clearly enough, it is worth while to eliminate from it the particles deviating largely from the average size. This is effected by the aid of an appliance used in the diamond trade for sorting cut stones according to size. The device consists of a metal receptacle in two parts separated by a plate pierced by a set of circular holes. On placing the

CHRISTIANSEN EXPERIMENT WITH SPHERICAL PARTICLES

material in the upper chamber and shaking it gently, the particles which are smaller than the rest fall into the lower chamber very quickly. The particles which are larger than the rest, on the other hand, remain in the upper chamber even after prolonged shaking. The particles which are either larger or smaller can thus be separated out from the middle fraction consisting of spherical particles having the same diameter as the circular holes in the sorter, viz., one millimetre. When a cleaning up of the spheres is found to be necessary, it is readily effected by washing with strong acids and subsequently with distilled water and a final drying.

2. Some optical consequences of the spherical shape

Acetone and carbon disulphide, the two liquids used, have refractive indices respectively lower and higher than the glass for the whole range of the visible spectrum. Hence, when a cell one centimetre thick containing the spherules is filled up with either one or the other liquid by itself, the medium is incapable of regularly transmitting an incident light beam. Nevertheless, a good deal of light does find its way through the cell in these circumstances. There is, however, a remarkable difference in its appearance in the two cases. When the spherules are surrounded by a liquid of lower index, the cell presents a brilliant sparkling appearance, due evidently to the emergence of light beams of considerable intensity from localised areas on its surface. The addition of a little carbon disulphide, though insufficient to render the cell transparent to any part of the spectrum, enhances the sparkling effect and makes it more attractive by reason of a play of colours similar to the "fire" of a diamond. On the other hand, when the liquid surrounding the spherules has a higher refractive index, these effects are not observed. The emergent light is then faint and diffuse, and the cell presents a dull appearance. The striking difference between the two cases, as well as the intermediate stages in which the cell is transparent to particular regions of the spectrum, can all be simultaneously observed by pouring enough carbon disulphide to fill the lower half of the cell, and then adding acetone to fill the upper half. The acetone, being lighter, floats above the carbon disulphide and interdiffusion takes place only slowly. The region of mixing appears as a bright band of transmitted colours, violet above and red below, while the upper and lower parts of the cell exhibit the effects arising from a penetration of the light through the cell without regular transmission.

An explanation in general terms may be readily given of the effects described above. A solid sphere surrounded by a liquid of lower index has a converging effect on a beam of light passing through it. *Per contra*, a light beam is made divergent by the sphere, when the surrounding liquid has a higher index. The effect of passage of the light through a series of spheres would accordingly be very different in the two cases. In one case, there would be a high probability that the light would emerge from the cell with its course deviated but nevertheless having

299

C V RAMAN: OPTICS

great concentrations of intensity in particular directions or at particular points on the surface of the cell. In the second case, the emergent beam would be strongly divergent and hence weak and diffuse at all points and in all directions. The correctness of this explanation can be checked by viewing a small bright source of light through the cell held in front of the eye. When the refractive index of the liquid is distinctly lower than that for the glass, the field of view surrounding the source exhibits a small number of bright spots drawn out into spectra irregularly distributed over its area. On the other hand, when the refractive index of the liquid is higher than that of glass, we observe an extended and diffuse halo exhibiting no noticeable detail.

The spherical shape of the particles has also other consequences. The deviation in the path of a ray of light by an individual sphere in the circumstances of the experiment would evidently be very small except when the ray is incident almost tangentially on the surface. Reflections of the light at the boundaries between the two media would also be of negligible intensity except in similar circumstances. Thus, the edges of the individual spheres grazed by the incident rays play a special role in determining the observed phenomena. When the cell is viewed directly, they show up as dark circles on a bright background. On the other hand, when the cell is viewed obliquely, the edges appear as bright crescents of light on a dark background. The convex parts of the edges then exhibit the colours for which the liquid has the higher refractive index, while the concave parts exhibit the remaining spectral colours. Hence, when there is an equality of index at or near one end of the visible spectrum, the concave or the convex parts alone (as the case may be) of the edges appear luminous when viewed obliquely, the other parts remaining dark.

4. The structure of the halo

In the foregoing, for the sake of simplicity, we have used the language of geometrical optics in describing and interpreting the observed phenomena. Nevertheless, as was stressed in a recent paper in these *Proceedings*, geometric theory is wholly inadequate as a basis for an understanding of the Christiansen effect. For, we are here principally concerned with regions in the spectrum for which the particles and the surrounding liquid have nearly identical refractive indices. The smaller the difference between these indices, the smaller would be the path-difference between the light rays traversing the cell along different routes and hence the more perfectly coherent would they be. Such coherence and the resulting regular transmission is the very essence of the Christiansen effect, and no explanation of the latter is therefore possible except on the basis of wave-optical considerations.

A study of the structure of the halo surrounding the light-source is very instructive in relation to the foregoing remarks. Remarkable changes are found to

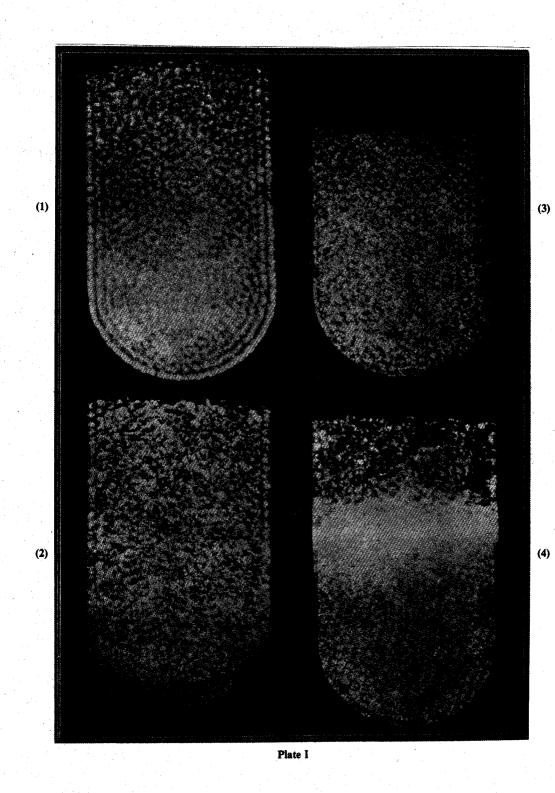
CHRISTIANSEN EXPERIMENT WITH SPHERICAL PARTICLES

occur in the halo when by progressive additions of carbon disulphide, the refractive index of the liquid is brought into coincidence with that of the spherules successively for different parts of the spectrum. The observations may be made with a small bright source of white light or alternatively with monochromatic radiation from a mercury or sodium lamp. In the former case, the part of the spectrum for which there is equality of refractive index disappears from the coloured streaks of light seen in the field of view and appears instead in the position of the light source. Observations with a monochromatic source reveal corresponding changes in the structure of the halo. The bright spots seen in the field when the liquid has a decidedly lower index are replaced by a complicated pattern of interference streaks and then by a halo with a mottled structure consisting of a great many diffraction images of the light source. This halo contracts in area and finally vanishes when the cell becomes optically homogeneous for the wavelength employed. The regularly transmitted light which builds up in intensity during the latter stages is then at its maximum. With further additions of carbon disulphide, it diminishes in intensity and finally disappears. Simultaneously, the halo reappears and increases in its angular extension until it covers a large area and is so diffuse that no detail can be observed in it.

5. The location of the spherules

When the spherules of glass are placed in the cell, they naturally arrange themselves in such manner as to occupy the smallest possible volume. But the walls of the cell necessarily influence and largely determine this arrangement. For, they fix the positions of the spheres actually in contact with them and therefore also, indirectly, the positions of the spheres further out in the interior of the cell. The Christiansen experiment enables us readily to demonstrate this. For, the transparency of the cell permits us to locate the spherules, and observe their positions with respect to each other and with respect to the walls of the cell. Indeed, the influence of the shape of the cell on the disposition of the particles in its interior is strikingly evident in the experiment. The layer of spherules in immediate contact with the walls naturally runs parallel to them. But their influence is observed to extend much further into the interior. At least four layers of particles running parallel to the walls of the cell may be clearly seen. This is irrespective of whether these walls are plane or curved or whether they are horizontal or vertical. When a rectangular cell is employed, five sets of such layers are observed running respectively parallel to the four walls and to the base of the cell and intersecting each other sharply along the edges of the cell.

The observations reported in the present paper are obviously of a qualitative character. It is felt however, that they are of sufficient interest to justify publication. We are engaged in a more detailed study of various aspects of the



CHRISTIANSEN EXPERIMENT WITH SPHERICAL PARTICLES

experiment and hope to be able to follow up the present communication by further papers.

6. Summary

Glass in the form of tiny spheres is technically available, and by the use of a suitable appliance, the material can be sorted out so as to consist of particles of uniform size. It can then be very effectively used to exhibit the Christiansen effect. The spherical shape of the particles as well as their uniformity of size have some striking consequences which are not observed in the usual form of the experiment in which an irregular pack of powdered glass is employed. Four photographs in plate I illustrate the paper.

Plate I. The photographs in plate I illustrate the appearance of the cell in different circumstances. They represent a threefold enlargement of its actual size. **Figure 1** exhibits the spontaneous arrangement of the spherules in layers parallel to the cell walls. Four such layers can easily be seen. The photograph was taken by transmitted light near the red end of the spectrum. **Figure 2** exhibits the appearance of the cell when filled with acetone and held against a bright source of light. **Figure 3** similarly exhibits its appearance when filled with carbon disulphide. Notice the numerous dark circles which represent the edges of the spheres. **Figure 4** illustrates the effect of filling the cell with carbon disulphide below and acetone above. See the text of the paper for further details of the effects observed in this case.