

THE ANOMALOUS DEPOLARISATION OF LIGHT-SCATTERING IN OPTICAL GLASSES.

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1. Introduction.

It is now well known that optical glasses exhibit a characteristic scattering of light, comparable in intensity with that exhibited by an ordinary dust-free liquid; in many cases, the Tyndall cone is of a beautiful sky blue colour and nearly, but not quite completely polarised when viewed in a transverse direction. Lord Rayleigh (1919) who remarked on the phenomenon, was inclined to attribute it to spherical inclusions in the glass of diameter not small, compared with the wave-length of light and explained the finite depolarisation as due to the appreciable size of these inclusions. In his investigations on the scattering of light in solid and fluid media, Prof. Raman (1922) took a different view, namely that the scattering in optical glasses arises fundamentally from the nature of the amorphous state of matter. He pointed out that we should expect two distinct types of scattering in amorphous solids, their relative importance depending on the particular circumstances of the case. The first type of scattering would result from the local (quasipermanent) fluctuations of optical density due to the non-uniform structure of the vitreous solid which differentiate it from the crystal; this type of scattering would, in respect of its intensity, be of the same order of magnitude as that arising from the (dynamic) fluctuations of optical density in the liquid at the temperature of fusion of the material. The second type of scattering would be due to the thermal agitation in the amorphous solid at the temperature of observation, and its intensity would be of the same order of magnitude as the light scattered in a crystal. In a later paper (Raman, 1927), he reported measurements of intensity and depolarisation of the light scattered transversely by a series of optical glasses, from which it appeared that low intensities and large depolarisations usually go hand in hand with low refractive indices, while glasses of high refractive indices generally give greater intensities and lower depolarisation. These results supported the view that the scattering is an intrinsic property of the glass. Very recently the author (Krishnan, 1936) made a comparative study of the light transversely scattered by seventeen different glasses of optical quality with the incident light in different states of polarisation, namely

unpolarised, vertically polarised and horizontally polarised. In all the cases studied, it is found that the values of the depolarisation factors ρ_u and ρ_v , corresponding to incident unpolarised light and light polarised with vibrations vertical respectively, are of the same order of magnitude as is usually observed in ordinary gases. On the other hand, when the incident light is polarised with vibrations horizontal, the horizontal component of the light scattered by the glass in the transverse horizontal direction, instead of being equal to the vertical component as in an ordinary gas or liquid, is definitely brighter than the vertical component, *i.e.*, the depolarisation factor ρ_h is less than unity as in the case of emulsions and colloids containing particles whose size is not small compared with the wave-length of light. This anomalous depolarisation* has been explained by the author as due to the existence of molecular aggregates in glasses, of size not small compared with the wave-length of the incident light. The composition of the glass is found to have a marked influence on the formation and the size of these molecular aggregates.

In a paper on the theory of the scattering of light, Hans Mueller (1938) has discussed the author's investigations on glasses in great detail. He has proposed a new theory of the scattering of light in glasses, based on the assumption that glasses contain a random distribution of frozen-in-strains. Slightly below the solidification temperature these strains are normal ones, but at lower temperatures shearing strains are created due to temperature contraction. It is remarkable to find that the magnitude of ρ_h as observed by the author follows very closely the order predicted by the theory. Mueller's paper serves to emphasise the fundamental character of the anomalous depolarisation (*i.e.*, ρ_h is less than 1), in relation to the study of the structure of amorphous substances.

Anomalous depolarisation is easily to be noticed in ordinary colloids, but is rather difficult to observe in optical glasses since the value of ρ_h is not far different from unity. The observation is rendered the more difficult by the feeble intensity of the light scattered in the transverse horizontal direction when the incident light is polarised with vibrations horizontal, and also by the feeble fluorescence exhibited by the optical glasses. Consequently in the earlier investigations, no attempt was made by the author to demonstrate the existence of anomalous depolarisation in an objective manner

* Any value of ρ_h less than unity is termed as the anomalous depolarisation. ρ_h is defined as the ratio of the intensity of the vertical to that of the horizontal component of the scattered light when the incident beam is polarised with vibrations horizontal.

as distinct from purely subjective (visual) measurements. But in view of the importance of the subject in relation to Mueller's theory of glass structure, it was thought desirable to have an objective demonstration of anomalous depolarisation in optical glasses.

2. *Experimental Details and Results.*

Two methods both based on photographic photometry are available for detecting the existence of anomalous depolarisation in any scattering medium. In the first method the transversely scattered light is photographed with the aid of two double-image prisms, one in the path of the incident beam and the other in the path of the scattered light. Of the four tracks thus obtained on the negative, corresponding to the four components V_v , H_v , V_h and H_h ,[†] the two middle ones should always be equal. If the fourth component H_h which is the horizontal component of the scattered light arising from the horizontal component of the incident beam is found to be distinctly brighter than either of the two middle anisotropic components, ρ_h will be greater than unity, from which it can be inferred that the optical heterogeneity of the scattering medium cannot be identified as due to the individual molecules, but due to small groups of molecules or due to the "frozen-in-strains" as Mueller has suggested. This method is not suitable for the case of optical glasses for the following reasons. The first component V_v is many times more intense than the other components. In consequence, the first component becomes enormously overexposed before a measurable record of the remaining three is obtained. The photographic halation produced thereby affects the intensity of the neighbouring components in such a way as to upset the equality of the middle two components and to equalise the last two components, namely, V_h and H_h , *i.e.*, to make ρ_h tend to unity. The detection of the small difference in the intensities of the two components V_h and H_h is thus rendered difficult.

The second method which is employed in the present investigation consists in photographing the scattered light through a double-image prism using incident polarised light. A narrow beam of light of square section was passed through the glass under investigation, the incident light being polarised by means of a square-ended nicol. The light scattered transversely in the horizontal direction was observed through a double-image prism which was kept suitably orientated so as to separate the vertical and the horizontal components of the scattered light in a vertical plane. Photographs of the

[†] These symbols have the same significance as those given in *Proc. Ind. Acad. Sci.*, (A), 1938, 1, 782.

two tracks were taken on the same negative for five different orientations of the nicol about the mean horizontal position. The corresponding nicol readings in degrees were 208, 203, 201, 199 and 194, where the reading 201 corresponded to that position of the nicol for which the incident beam was polarised with vibrations horizontal. Each negative containing the five pairs of tracks was passed through a Moll microphotometer and the intensities of the two tracks in each pair were compared. Three glasses having melt numbers 23975, 23497 and 22608 and also a bulb containing dust-free ethyl ether were examined under identical conditions. The respective microphotometric records are reproduced in Figs. 1 to 4.

For the glass 23975 when the nicol reading is 208 the vertical component of the scattered light is definitely brighter than the horizontal component (see Fig. 1). As the nicol is rotated towards the horizontal position, the two components become equal in intensity and when the reading is 201, *i.e.*, when the nicol is in the exact horizontal position, the horizontal component is definitely brighter than the vertical component thus giving rise to a value of ρ_h less than unity. When the nicol is rotated further in the same direction, the two components of the scattered light again become equal in intensity and for a further rotation of the nicol the vertical component becomes brighter than the horizontal component. The reversal of intensities of the two components is clearly seen in Figs. 2 and 3. But it is not so very conspicuous as in Fig. 1. This is what should be expected since the observed values of ρ_h for these two glasses are higher than that for glass 23975. The following table gives the comparative values of ρ_u , ρ_v and ρ_h for the three glasses and also for ether.

TABLE I.

Scattering substance	ρ_u^*	ρ_v^*	ρ_h^*
Glass No. 23975	0.012	0.0046	0.68
Glass No. 23497	0.045	0.02	0.77
Glass No. 22608	0.061	0.03	0.91
Ethyl ether	0.08	0.042	1.00

* These values are taken from *Proc. Ind. Acad. Sci.*, (A), 1936, 3, 216.

The microphotometric record taken with ether does not exhibit any reversal in the relative brightness of the two components. For nicol readings

H ↑ V ↑ H ↑ V ↑ H ↑ V ↑

FIG. 1.—Glass No. 23975

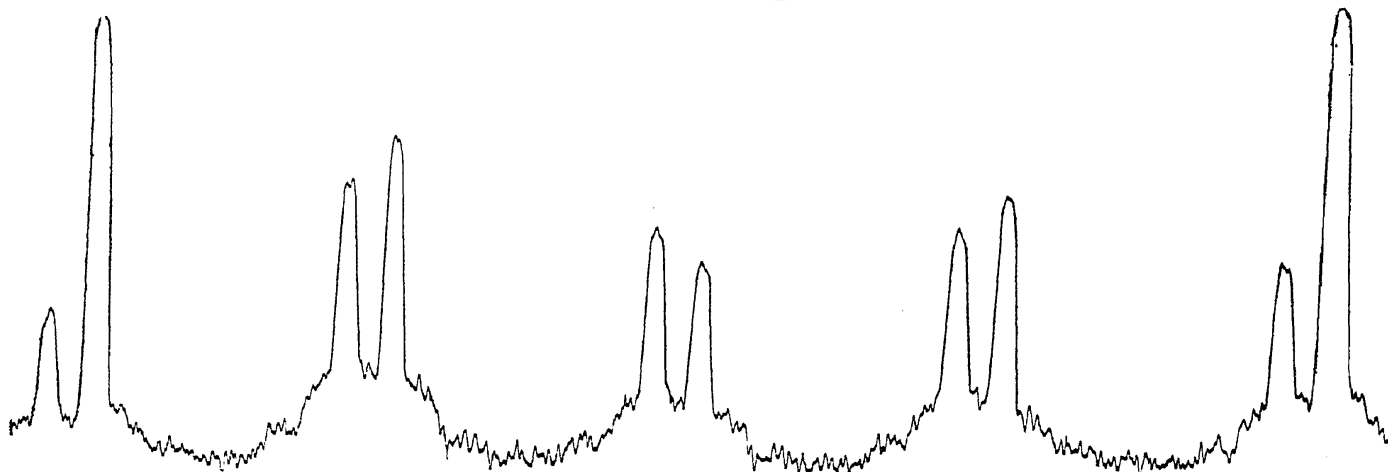


FIG. 2.—Glass No. 23497

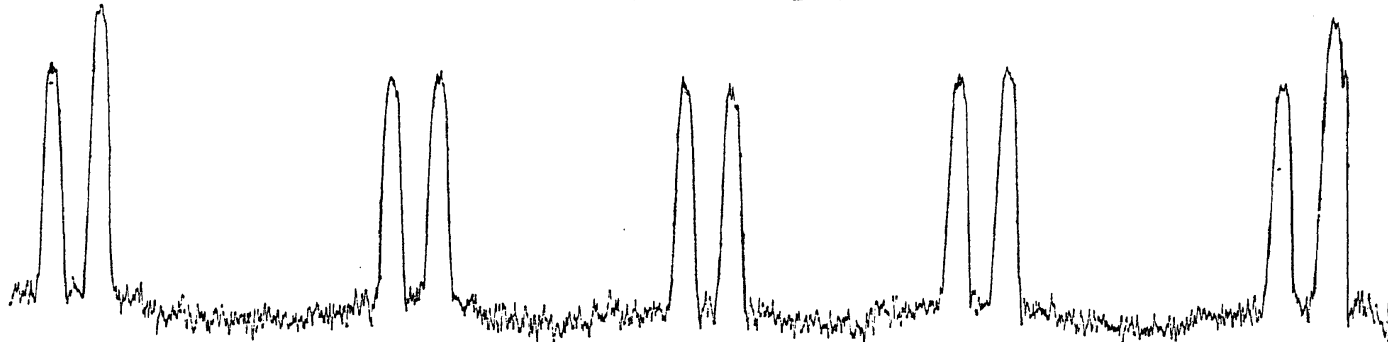


FIG. 3.—Glass No. 22608

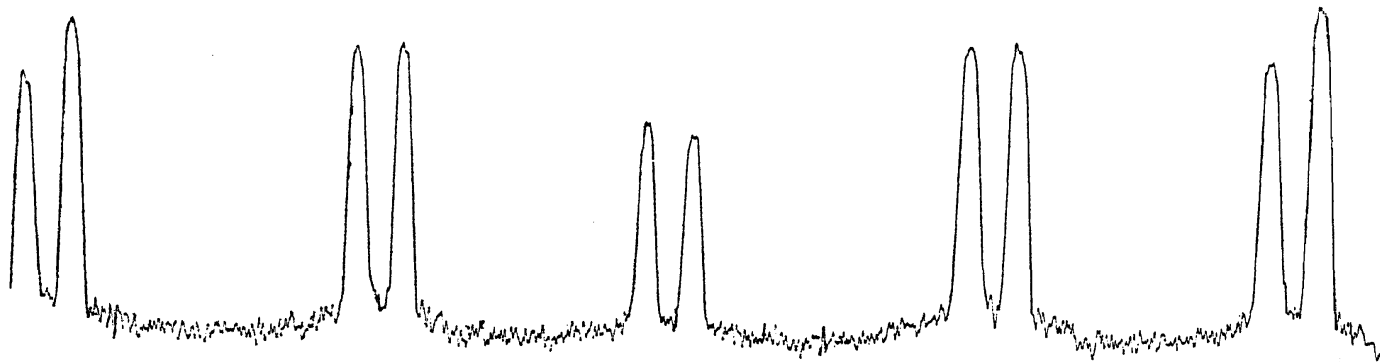
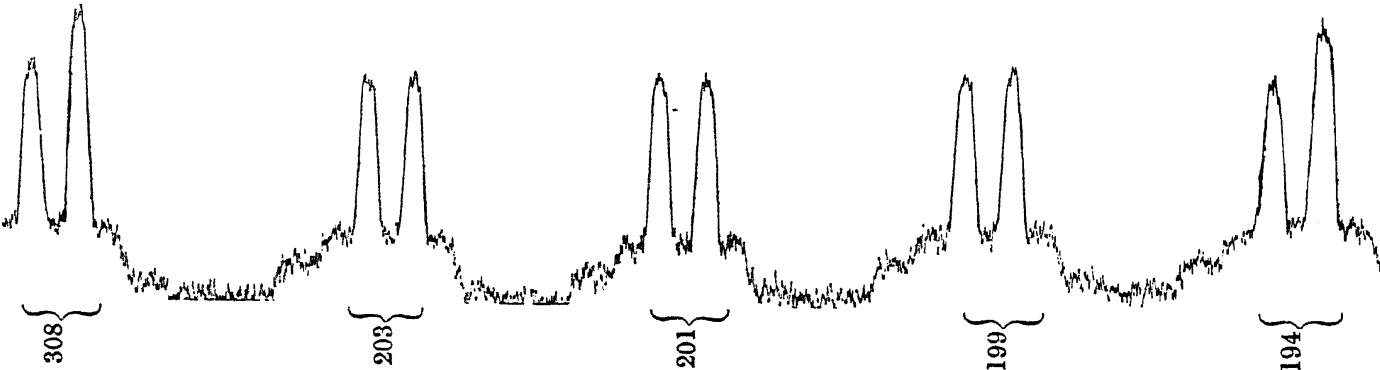


FIG. 4.—Ether



Nicol readings

194, 199, 203 and 208 the vertical component is definitely brighter than the horizontal component, whereas for nicol reading 201 the two are equal in intensity showing thereby that ρ_h has the limiting value of unity. This is in accordance with the theoretical considerations. From this it can be inferred that the anomalous depolarisation observed in the case of the three optical glasses investigated is a genuine effect and not due to any defect in the experimental technique. If suitable coloured filters are used to eliminate fluorescence, the reversal in the relative brightness of the two components can be seen more conspicuously.

In conclusion, the author takes this opportunity to express his grateful thanks to Prof. Sir C. V. Raman for his kind and helpful interest in the work.

3. Summary.

An experimental method based on photographic photometry is described for demonstrating the existence of anomalous depolarisation, *i.e.*, ρ_h less than unity, in optical glasses. Microphotometric records are reproduced which clearly show the existence of anomalous depolarisation in optical glasses. Three typical glasses are examined. The experiment was repeated with ether as the scattering substance and a negative result was obtained showing thereby that there was no defect in the experimental technique.

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