ON THE CONVERGENCE ERROR IN DEPOLARISATION MEASUREMENTS.

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1. Introduction.

So much importance attaches to the precise measurement and interpretation of the degree of depolarisation of scattered light—be it in Tyndall, Rayleigh or Raman scattering—that no apology is needed for a paper dealing with a somewhat difficult point arising in the technique of such measurements, namely, the errors due to the lack of parallelism in the rays of light entering the medium and scattered by it. In practice it is not possible to realise the theoretical ideal of the illumination of the medium by a beam of parallel rays, and the examination of the scattered light in a direction strictly transverse to the latter. The comparatively feeble intensity of the scattered light usually entails the use of a source of finite dimensions and of a lens to concentrate the beam of light during its passage through the medium. In consequence the rays of light are neither parallel among themselves, nor strictly perpendicular to the direction of observation, and errors arise which may assume great importance when considering relatively small depolarisations.

There has been considerable divergence of opinion regarding the magnitude of the errors arising in the manner referred to above, and particularly in the case when an illuminating lens of large aperture is used to focus an image of the source of light within the medium at the point of observation. It is obvious that the rays starting from a given point on the source and reaching the conjugate point in the focal plane are optically coherent with each other. Dr. I. Ramakrishna Rao¹ took this coherence into account and claimed that no correction is necessary to the observed value of the depolarisation on account of the finite aperture of the lens provided the measurements are made precisely at the focus. On the other hand Gans² assumed the rays meeting at the focus to be incoherent—an assumption

obviously not justifiable—and derived the formula
\[ \rho = \rho_0 + \frac{1}{2} \sin^2 \theta = \rho_0 + \frac{\theta^2}{2} \]
where \( \rho_0 \) = depolarisation with parallel unpolarised light
and \( \rho \) = observed value of the depolarisation when the incident beam
has a semi-convergence \( \theta \).

Cabannes\(^3\) has discussed these rival views in his book and attempted to
reconcile them, but the question has remained unsettled and somewhat
obscure.

2. Case of two Intersecting Parallel Beams.

To elucidate the points at issue it is desirable to treat at first a relatively
simple case.

Consider a pair of parallel, coplanar, plane polarised beams crossing each
other at an angle \( 2 \theta \). Let the electric vector in the beams be in the plane
of the paper which is supposed to be the xy-plane. The region over which
the two beams are superposed will be an interference field, the interference
maxima and minima being bands parallel to the x-axis, occurring with a
regular periodicity \( \frac{\lambda}{4 \sin \theta} \).

![Fig. 1.](image)

At the interference maxima (represented by thick lines in the figure), the
x-components of the electric intensity annul each other so that the resultant
electric vector is vertical, while the interference minima (represented by
dotted lines in the figure) are places where the resultant vibration is parallel.

\(^3\) J. Cabannes, \textit{La Diffusion Moleculaire de la Lumiere}. 
to the axis of $x$. If the region of overlapping is occupied by molecules of a gas which scatter light in the usual way, and if one were to make observations in a direction perpendicular to the plane of the paper, the depolarisation observed along the interference maxima will be the correct value, while at the interference minima, the electric vector being horizontal the depolarisation ratio will be reversed. Let it now be assumed that on account of some reason, such as for instance, the lack of monochromatism in the interfering beams or the excessive closeness of the interference bands, that it is not possible to concentrate attention on the individual interference maxima. The depolarisation measured in the usual way in such circumstances would be that averaged over the whole field, due regard being had to the intensity, and would thus differ from the correct value.

At any point $(x, y)$ in the interference field where the relative retardation between the interfering beams is $\delta = 2y \sin \theta$, the components of the electric intensity are given by

$$
E_x = 2E \sin \theta \sin (k y \sin \theta) \sin (vt + y \sin \theta),
$$
$$
E_y = 2E \cos \theta \cos (k y \sin \theta) \cos (vt + y \sin \theta),
$$
$$
E_z = 0
$$

where $k = \frac{2\pi}{\lambda}$

The time mean square of the components of the electric moment induced in a molecule situated at $x, y$ are given by the equations

$$
\overline{p_x^2} = (A - B) \overline{E_x^2} + B (\overline{E_x^2} + \overline{E_y^2} + \overline{E_z^2})
$$
$$
\overline{p_y^2} = (A - B) \overline{E_y^2} + B (\overline{E_x^2} + \overline{E_y^2} + \overline{E_z^2})
$$
$$
\overline{p_z^2} = (A - B) \overline{E_z^2} + B (\overline{E_x^2} + \overline{E_y^2} + \overline{E_z^2})
$$

where $A = \frac{1}{5} \sum g_1^2 + \frac{2}{15} \sum g_1 g_2$

and $B = \frac{1}{15} (\sum g_1^2 - \sum g_1 g_2)$

$g_1, g_2, g_3$, being the principal polarisabilities of the molecule.

The components of the intensity of the scattered radiation polarised parallel to the $x, y$ and $z$ directions will be obtained by integrating (2) over the entire field. If we call these components $P_x^2, P_y^2, P_z^2$, then the depolarisation for observation in a direction perpendicular to the plane of the paper is given by

$$
\rho = \frac{P_x^2}{P_y^2} = \frac{A \sin^2 \theta + B \cos^2 \theta}{B \sin^2 \theta + A \cos^2 \theta} = \rho_0 + \theta^2
$$

where $\rho_0$ is the depolarisation when $\theta = 0$.

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4 R. Cans, Loc. cit.
The same result follows if the two beams are considered as incoherent and if we adopt the procedure of Gans.

3. Lens with Square Aperture.

We shall next consider the case in which a lens of focal length $f$ limited by a square aperture of edge $a$ is used to concentrate the light on the medium. Let the plane of the aperture be the $xy$-plane and let the direction of propagation be the $x$-axis. Let the incident light before its passage through the lens be polarised with the electric vector parallel to the axis of $x$, and let the vibrations be given by $E \cos kvt$.

The components of the electric intensity at any point in the focal plane whose co-ordinates are $\zeta$ and $\eta$ due to an element $dx \, dy$ of the aperture whose co-ordinates are $x, y, z$ are given by

$$
E_x = -\frac{1}{\lambda f} E \cos \phi \cdot \sin \frac{1}{\lambda f} \left( vt - f + \frac{ax + by}{f} \right) dx \, dy
$$

$$
E_y = 0
$$

$$
E_z = -\frac{1}{\lambda f} E \sin \phi \cdot \sin \frac{1}{\lambda f} \left( vt - f + \frac{ax + by}{f} \right) dx \, dy
$$

where $\phi$ is the inclination of the electric vector in the ray from the element with the axis of $x$.

As a first approximation we may put

$$
\cos \phi = 1 \quad \text{and} \quad \sin \phi = \frac{a}{f}
$$

The components of the electric intensity at $M$ due to the whole aperture are obtained by integrating (4) over the entire aperture. They are

$$
E_x = -\frac{E}{\lambda f} a^2 \sin \frac{1}{\lambda f} \left( vt - f \right) \left[ \frac{\sin u}{u} \cdot \frac{\sin v}{v} \right]
$$

$$
E_y = 0
$$

$$
E_z = -\frac{E}{\lambda f} a^2 \cos \frac{1}{\lambda f} \left( vt - f \right) \cdot \left[ \frac{\sin v}{v} \right] \left[ \frac{\sin u}{u^2} - \frac{\cos u}{u} \right]
$$

where $u = \frac{\pi a^2}{f \lambda}$ and $v = \frac{\pi a^2}{f \lambda}$.

Substituting these values of $E_x, E_y, E_z$ in equations (2) of the preceding section, and following the same treatment we get, after putting $\frac{a}{f} = 2\theta$

$$
\bar{P}_x^2 = \left. \int \int (A \bar{E}_x^2 + B \bar{E}_x \bar{E}_y) d\zeta \, d\eta \right|_{-\infty}^{+\infty} = \frac{1}{2} E^2 a^2 [A + B \frac{\theta^2}{2}]
$$

$$
\bar{P}_y^2 = \left. \int \int (B \bar{E}_x^2 + B \bar{E}_y \bar{E}_z) d\zeta \, d\eta \right|_{-\infty}^{+\infty} = \frac{1}{2} E^2 a^2 [B + B \frac{\theta^2}{2}]
$$

$$
\bar{P}_z^2 = \left. \int \int (B \bar{E}_x \bar{E}_y + A \bar{E}_z^2) d\zeta \, d\eta \right|_{-\infty}^{+\infty} = \frac{1}{2} E^2 a^2 [B + A \frac{\theta^2}{2}]
$$

From these it follows that the depolarisation of the light scattered in the
direction $Oy$ is given by
\[ p_{\nu} = \frac{P_{1}^2}{P_{0}^2} = \rho_{\nu 0} + \frac{1}{3} \theta^2 \ldots \ldots \ldots \ldots \ldots \ldots \ldots (7) \]
where $\rho_{\nu 0}$ is the value of $\rho_{\nu}$ for $\theta = 0$.

If observations are made in the direction $Ox$, we have
\[ p_{\nu} = \frac{P_{2}^2}{P_{y}^2} = 1 + \frac{1}{\rho_{\nu 0}} \frac{1}{3} \theta^2 \ldots \ldots \ldots \ldots \ldots \ldots \ldots (8) \]

If the incident light is unpolarised, it is easily seen that the depolarisation
of the transversely scattered light is given by
\[ p_{\nu} = \rho_{\nu 0} + \frac{1}{3} \theta^2 \ldots \ldots \ldots \ldots \ldots \ldots \ldots (9) \]

From (7) and (9) it follows that so far as convergence error is concerned,
no advantage is gained by using light polarised with the electric vector
vertical in place of unpolarised light, since $\rho_{\nu 0}$ is nearly half of $\rho_{\nu 0}$.

From (a) we see that when the incident light is polarised with the electric vector
horizontal, the effect of convergence is to make the horizontal component
appear brighter than the vertical and this effect becomes all the more striking
when the value of the depolarisation is very small. Formulae (7), (8) and
(9) also follow readily if the incident beam is treated as incoherent, and if
we adopt the method of Gans. What actually takes place is that the finite
width of the source such as the sun, for instance, produces a mixing up of the
diffraction patterns due to the different points of the source so that the ultimate
effect is the same as though the rays from any single point are incoherent.

4. Lens with Circular Aperture.

We now go to consider the case of greater practical interest and impor-
tance, namely, the case in which the condensing lens is limited by a circular
aperture of radius $R$.

We follow the treatment already given for the square aperture. Putting
\[ k_x = \phi \text{ and } k_y = \eta, \]
and noting that the form of the aperture is sym-
metrical with respect to the axes of $x$ and $y$, the expressions for the electric
intensity at any point in the focal plane may be written in the form
\[ \begin{align*}
E_x &= -\frac{E}{\lambda f} \sin \phi (vt - f) \int \cos \phi x \cos \eta y \; dx \; dy \\
E_y &= 0 \\
E_z &= \frac{E}{\lambda f^2} \cos \phi (vt - f) \frac{d}{dp} \left[ \int \cos \phi x \cos \eta y \; dx \; dy \right]
\end{align*} \ldots (10) \]

When the convergence of the incident beam is not too great, it is justi-
fiable as a first approximation to assume that the $x$-component of the electric
intensity is symmetrical with respect to the focal point $\phi = 0$, $\eta = 0$. It is
thus sufficient to determine $E_x$ along the axis of $\phi$, and so the integral in (10)
reduces to the usual diffraction integral in the case of the circular aperture. We thus obtain for the time mean square of $E_x$, $E_y$ and $E_z$
\[
\begin{align*}
\langle E_x^2 \rangle &= \frac{E^2}{2\lambda f^2} \left[ \frac{2J_1 \left( \frac{2\pi Rr}{f\lambda} \right)}{\left( \frac{2\pi Rr}{f\lambda} \right)^2} \right]^2 \\
\langle E_y^2 \rangle &= 0 \\
\langle E_z^2 \rangle &= \frac{E^2}{2\lambda f^2} \left[ \frac{d}{d\phi} \left( \frac{2J_1 \left( \frac{2\pi Rr}{f\lambda} \right)}{\left( \frac{2\pi Rr}{f\lambda} \right)^2} \right) \right]^2
\end{align*}
\] 

where $r$ is the distance of the point in the focal plane under consideration from the focus $\rho = 0$, $q = 0$.

It is readily shown that
\[
\int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \langle E_x^2 \rangle \, d\xi \, d\eta = \frac{E^2}{2} \pi R^2 2 \int_{0}^{\infty} z^{-1} J_1^2 (z) \, dz = \frac{E^2}{2} \pi R^2 
\]
and
\[
\int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \langle E_z^2 \rangle \, d\xi \, d\eta = \frac{E^2}{2} \pi R^2 \frac{R^2}{f^2} \int_{0}^{\infty} z^{-1} J_2^2 (z) \, dz = \frac{E^2}{2} \pi R^2 \frac{4\theta^2}{f^2} 
\]

where $z = \frac{2\pi Rr}{f\lambda}$ and $\theta = \frac{R}{f}$.

Proceeding to find $\rho_\alpha$, $\rho_\beta$ and $\rho_\gamma$, as in the case of the square aperture we obtain,
\[
\begin{align*}
\rho_\alpha &= \rho_{\alpha 0} + \frac{1}{4} \theta^2 \\
\rho_\beta &= 1 + \frac{1}{\rho_{\alpha 0}} \frac{1}{4} \theta^2 \\
\rho_\gamma &= \rho_{\gamma 0} + \frac{1}{4} \theta^2
\end{align*}
\] 

The last of the results (14) has been worked out by Gans using the idea of incoherence, while the first two which relate to the cases in which the incident light is plane-polarised can be obtained by adopting the same procedure. The significance of (14) has been already explained in section (3).

5. Remarks on the Foregoing Results.

The above investigation relates to the focal plane and assumes that the lens is uniformly illuminated and that every part of the aperture is equally effective at any given point in the focal plane where there is any appreciable intensity. This requires the use of a lens which is optically perfect. Whether it is so or not can be determined by the well-known knife-edge test. Sunlight reflected by a heliostat is allowed to illumine the entire aperture of the lens, and the rays diverging from the focus are allowed to fall on a distant screen. A sharp razor-blade is slowly made to advance through the focal
plane and the illumination of the circular patch of light on the screen is watched. If the lens is optically good, the illumination of the entire area fades off quite regularly and uniformly, so that by merely looking at the screen it is impossible to find out from which side the knife-edge is cutting across. Actual experiment with a high-class photographic lens shows that even for a quite appreciable distance on either side of the focal plane, the knife-edge test gives the same result, and within this region, the theoretical results as regards the magnitude of the convergence correction would be expected to be very nearly valid. As the extent of this region is quite comparable with the dimensions of the track over which one concentrates in practice in depolarisation work, the theoretical correction can be safely applied to the observed value so long as the lens used in the experiment is optically good, and observations are restricted to the vicinity of the focus. When we deviate far too much from the focal plane, the knife-edge test reveals a decided asymmetry in the illumination at any point, which is shown by the definite appearance of the shadow of the knife-edge marching across the illuminated area on the screen. Under such circumstances the validity of the theoretical treatment would no longer hold good, and the magnitude of the correction at any point will depend upon whether it receives illumination from the central or marginal portions of the lens. The actual correction will be smaller than the theoretical value in the former case, and larger in the latter, and the observed depolarisation of the transversely scattered light will thus vary from point to point within the visible track of the illuminating beam within the medium.

So far the necessity for the use of an optically perfect lens has been stressed as a necessary consequence of the theoretical considerations, and this evidently implies that it is also of the highest importance to have the rest of the optical parts as perfect as possible. If sunlight is employed for depolarisation work, it is essential to use a good heliostat mirror to reflect the light into the observation chamber. To avoid distortion of the incident beam as it enters the vessel containing the scattering medium, it is essential to use a flat window. The use of a bulb is highly unsatisfactory in this respect. It is desirable also that the window through which the scattered track is viewed should be free from optical defects.

6. Some Experimental Results.

It has been already remarked that the convergence error becomes very important when the genuine depolarisation of the substance under investigation is very small. This at once suggests that gases and vapours of small depolarisation as well as liquids and liquid mixtures near the critical point are the media in which the depolarisation would be expected to be most
susceptible to the change in convergence of the incident beam. In a practical point of view, however, the intensity of scattering is an important factor in the experimental investigation of the question; accurate determination of the depolarisation over a wide range of convergence is feasible only if the intensity of scattering is relatively large.

(a) Gases.—Isobutane seemed to be favourable in the case as its depolarisation is very small, while the scattering is at the same time relatively intense. The gas was taken from a cylinder supplied by Ohio Chemical Works, and was guaranteed to be 99% pure. The depolarisation was determined independently using two different arrangements:

(1) The gas was contained in a pear-shaped bulb of the type used by Ramanathan and I. R. Rao in their investigations, but with the addition of a projecting tube with flat window on the observation end. The light was reflected by a Foucault heliostat was concentrated at the centre of the screen by using a high class condensing lens in combination with an adjustable diaphragm, and the depolarisation of the transversely scattered light was measured in the usual way. The angle of convergence of the incident beam was determined by measuring the diameter of the illuminated screen placed at a distance of one metre from the focus. The results are given in the table below:

<table>
<thead>
<tr>
<th>Convergence of the incident beam = $2\theta$</th>
<th>Observed value of the depolarisation = $\rho_{\mu}$</th>
<th>Correction $\frac{\theta^2}{2}$</th>
<th>Corrected value of the depolarisation = $\rho_{\mu c}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Deg. Radian</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$29^\circ 48' = 0.520$</td>
<td>$4.04%$</td>
<td>$3.38%$</td>
<td>$0.66%$</td>
</tr>
<tr>
<td>$19^\circ 48' = 0.346$</td>
<td>$2.27%$</td>
<td>$1.45%$</td>
<td>$0.82%$</td>
</tr>
<tr>
<td>$9^\circ 54' = 0.173$</td>
<td>$1.06%$</td>
<td>$0.37%$</td>
<td>$0.69%$</td>
</tr>
</tbody>
</table>

Mean value = $0.72\%$

The values in the last column show that, in spite of the large observed depolarisation when the convergence of the incident beam is large, the corrected value comes out to be very nearly the same at small values of convergence.

(2) The gas was contained in a cross made of pyrex glass with end plates and suitable diaphragms. A Dallmeyer photograph
focal length 1 foot and adjustable aperture was used for concentrating the sunlight at the centre of the cross. The optics of the whole arrangement was thus much superior to that used in the first case. The results are recorded in the table below.

**Table II.**

<table>
<thead>
<tr>
<th>Aperture</th>
<th>$\rho_n$</th>
<th>$\frac{\theta^2}{2}$</th>
<th>$\rho_{n0} = \rho_n - \frac{\theta^2}{2}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>F/5.6</td>
<td>0.83%</td>
<td>0.4%</td>
<td>0.43%</td>
</tr>
<tr>
<td>F/11</td>
<td>0.58%</td>
<td>0.1%</td>
<td>0.48%</td>
</tr>
</tbody>
</table>

Mean value = 0.46%

The difference in the values of the depolarisation measured with the bulb and with the cross, emphasises the necessity for the perfection of the optical parts used in depolarisation work.

(b) **Liquids.**—A critical mixture of methyl alcohol and hexane distilled and sealed in a spherical bulb was found to be specially suited for the study of the convergence correction in the case of liquids, because of the close proximity of its critical temperature (29°C.) to the room temperature. The mixture was warmed just above the critical point of complete miscibility and was placed in the path of the incident beam in such a way that the centre of the bulb coincided with the focus. The use of a spherical bulb as stated above eliminates the alteration in the convergence of the track which would otherwise require to be considered. The observed and corrected values of the depolarisation are given below.

**Table III.**

<table>
<thead>
<tr>
<th>$2\theta$</th>
<th>$\rho_n$</th>
<th>$\frac{\theta^2}{2}$</th>
<th>$\rho_{n0} = \rho_n - \frac{\theta^2}{2}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Deg.</td>
<td>Radian</td>
<td></td>
<td></td>
</tr>
<tr>
<td>28° 20'</td>
<td>=0.4945</td>
<td>3.34%</td>
<td>3.05%</td>
</tr>
<tr>
<td>18° 11'</td>
<td>=0.3173</td>
<td>1.49%</td>
<td>1.26%</td>
</tr>
<tr>
<td>4° 35'</td>
<td>=0.08</td>
<td>0.44%</td>
<td>0.08%</td>
</tr>
</tbody>
</table>

Mean value = 0.29%

The observation of the Tyndall cone through a nicol in the case of the liquid mixture reveals certain interesting features which illustrate the
theoretical discussion contained in section (5). Using an incident beam of large convergence, when the nicol through which the transversely scattered light is viewed is rotated so as to transmit only the horizontal component, one finds a dark region in the middle of the Tyndall cone on either side of the focus, while the margins appear as a pair of bright streamers (Fig. 3 in the Plate). A slight rotation of the nicol either way makes the luminous cone asymmetric, the intensity fading off rapidly from one edge to the other (Figs. 2 and 4 in the Plate). The explanation for these appearances is almost self-evident in view of what has been stated in section (5). Fig. 3 in the Plate brings out in a picturesque fashion the whole mechanism of the convergence error. The dark centre shows how the genuine depolarisation is exceedingly small, and how the greater part of the illumination observed at the focus is contributed by the marginal portions of the lens.

In conclusion, it is my greatest pleasure to record my respectful thanks to Professor Sir C. V. Raman, for suggesting the present investigation, and for much valuable guidance and criticism in the course of the work.

7. Summary.

The scattering of light in an interference field is discussed, and it is shown that for the simple case of two parallel plane-polarised intersecting beams, the depolarisation at the interference maxima gives the correct value, while at the interference minima the depolarisation ratio is reversed. The average of the depolarisation taken over the whole field is higher than the correct value. The treatment is extended to the cases in which a lens covered with a square aperture, and with a circular aperture, respectively, is used to concentrate the light on the scattering medium. It is shown that the observed values of the depolarisation would deviate from the genuine values by a correction factor which involves the square of the angle of convergence. The observed depolarisation \( \rho \) is given by

\[
\begin{align*}
\rho_v &= \rho_{v0} + a\theta^2 \\
\rho_h &= 1 + \frac{1}{\rho_{v0}} a\theta^2 \\
\rho_u &= \rho_{u0} + 2a\theta^2
\end{align*}
\]

where \( a \) has the value \( \frac{1}{2} \) for a square aperture, and \( \frac{1}{2} \) for a circular one. The subscripts \( v, h, u \) refer to the cases in which the incident light has its electric vector vertical, horizontal and is unpolarised respectively. It is pointed out that the same results follow by treating the incident beam as a bundle of incoherent rays. Some consequences of the theoretical results are discussed and the necessity for the perfection of the optical parts used in depolarisation work is emphasised. Experimental results are given which illustrate the points discussed in the paper.
R. Ananthakrishnan.  