# OPTICAL THEORY OF CHROMATIC EMULSIONS AND OF THE CHRISTIANSEN EXPERIMENT

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## 1. *Introduction*

While engaged upon some determinations of the refractive index of transparent powders by the method of immersing them in liquid mixtures of the same refractive index, Christiansen (1884) observed some very remarkable and interesting colour effects. If white light was employed, the transmitted light was highly coloured, the colour corresponding to the particular wave-length for which the two substances have the same refractive index. Lord Rayleigh (1885) repeated these experiments, and found that the transmitted spectrum was remarkably narrow. The subject was investigated in detail by Sethi (1921), who studied the effect of the size and number of the particles and of the thickness of the medium on the transmitted light, and also the light scattered in other directions. He extended his studies to the colours shown by emulsions of immiscible liquids, whose refractive indices are equal for some wave-length in the visible range. The elegant method of preparing these emulsions in a stable state, discovered by Holmes and Cameron (1922), enabled Sogani (1926) to make a thorough study of the phenomena exhibited by these "chromatic emulsions".

Lord Rayleigh has rightly remarked (1899) that a proper theory of the Christiansen phenomenon must be based on wave-optical ideas. Following the lines suggested by Rayleigh, both Sethi and Sogani have tried to explain, theoretically, the phenomena observed by them. But their method is not comprehensive, since separate theories have to be worked out to explain the transmission colours, and the colours of the light scattered in other directions. In this paper, an attempt is made to explain the whole range of phenomena on a single theory based on the diffraction of light by a transparent sphere, whose refractive index is not appreciably different from that of the surrounding medium. The author (1943 *a*) has already developed such a theory in connection with the study of the transmission of light through a cloud of transparent droplets. As a particular case, the theory

has been used to explain the occurrence of coronæ when a bright object is viewed through thin cloud (1943 *b*).* The application of the theory to the explanation of the properties of chromatic emulsions forms another particular case, not covered by the previous one. The theory is strictly applicable only to spherical particles, and hence only to the case of chromatic emulsions; nevertheless as we shall see in the-course of the paper, it suffices to give a good account of the phenomena observed in the Christiansen experiment with particles of arbitrary shape.

## 2. *Intensity and Spectral Nature of the Transmitted Light*

If a sphere of radius $a$, and of refractive index $\mu$, be placed in a medium of refractive index $\mu_0$, then the amplitude of the wave diffracted in a direction making an angle $\phi$ with the incident direction can be shown (1943 *a*) to be

$$X = X_1 - X_2, \tag{1}$$

where
$$X_1 = K \sin \chi \int_0^{\pi/2} J_0 (\eta \sin \theta) \cos (\xi \cos \theta) \sin \theta \cos \theta \, d\theta$$
$$- K \cos \chi \int_0^{\pi/2} J_0 (\eta \sin \theta) \sin (\xi \cos \theta) \sin \theta \cos \theta \, d\theta, \tag{2}$$

$$X_2 = K \sin \chi \cdot J_1 (\eta)/\eta, \tag{3}$$

$$\xi = 4\pi (\mu - \mu_0) a/\lambda, \; \eta = 2\pi a \mu_0 \sin (\phi)/\lambda \text{ and } K = 2\pi a^2/\lambda, \tag{4}$$

the incident wave being represented by $\sin \chi$, and $\lambda$ being its wave-length in vacuum.

In the exact forward direction, however, the effect of the incident wave would also be present. If we denote the amplitude due to the incident wave as $X_0$, then that due to the transmitted wave is

$$X' = X_0 + X_1' - X_2' \tag{5}$$

where $X_1'$ and $X_2'$ are the values of $X_1$ and $X_2$ when $\phi = 0$. For this case, $\eta = 0$, and the integrals in (2) can be integrated (1943 *a*) giving

$$X' = X_0 + K \sin \chi \left\{ \frac{\cos \xi}{\xi^2} + \frac{\sin \xi}{\xi} - \frac{1}{\xi^2} - \frac{1}{2} \right\}$$
$$- K \cos \chi \left\{ \frac{\sin \xi}{\xi^2} - \frac{\cos \xi}{\xi} \right\}$$
$$= X_0 - KC_1' \sin \chi - KS' \cos \chi \text{ (say)} \tag{6}$$

* Hereafter, these papers will be referred to as (1943 *a*) and (1943 *b*) respectively.

where $K = 2\pi a^2/\lambda$. Now, if A be the aperture of the beam, $X_0$ is evidently $= A \sin(\chi)/\lambda$, so that

$$X' = \frac{A}{\lambda} \sin \chi \left\{1 - \frac{2\pi a^2}{A} C_1'\right\} - \frac{A}{\lambda} \cos \chi \left\{\frac{2\pi a^2}{A} S'\right\}. \tag{7}$$

The above expression applies to a single droplet. Applying the method used in Section 5 of (1943 *a*), it is easily shown that the attenuation coefficient is

$$b = -4\pi a^2 N C_1' \tag{8}$$

and the refractive index, $n$, is given by

$$n - \mu_0 = N\lambda a^2 S' \tag{9}$$

where N is the number of droplets per unit volume. Thus, if $l$ be the thickness of the emulsion, then the intensity of the transmitted light is

$$I = I_0 \exp[-bl] = I_0 \exp[-4\pi a^2 N l C_1'], \tag{10}$$

where $I_0$ is the intensity of the incident light.

The value of $C_1'$ is given by (6). But, in the Christiansen experiment, and in the case of chromatic emulsions, the refractive indices of the two media are the same for some particular wave-length, say $\lambda_0$, and differ at other wave-lengths. The important colour effects, and other phenomena in which we are interested occur in a small range of wave-lengths about $\lambda_0$, where $(\mu - \mu_0)$ has only a very small value. Hence, the range of values of $\xi$ that would be required will be only small, comprising of small positive and negative values. In order to be applicable for this range, $C_1'$ can be expanded in the form of a power series in $\xi$ as

$$C_1' = \xi^2/8 - \xi^4/144 + \cdots \tag{11}$$

so that, for small values of $\xi$, $C_1' = \xi^2/8$. If larger values of $\xi$ occur in any case, expression (6) can be used.

Hence, for small values of $\xi$,

$$I_l/I_0 = \exp[-\pi a^2 N l \xi^2/2]. \tag{12}$$

This equation gives the variation of the intensity of the transmitted light with various factors.

Taking first the variation with $\xi$, it is easily seen that $I_l = I_0$ at $\xi = 0$, and decreases rapidly as $\xi^2$ increases, the graph connecting $I_l$ and $\xi$ having the shape of the well-known probability curve. Now $\xi$ is a function of both $(\mu - \mu_0)$ and $\lambda$. But, the variation of $\xi$ with $\lambda$ is only a steady one, so that we shall at first neglect this variation, and consider only the effect of changes in the value of $(\mu - \mu_0)$. The effect of the change in the wave-length will be finally discussed.

Substituting the value for $\xi$, (12) becomes

$$I_l/I_0 = \exp\left[-8\pi^3 N l (\mu-\mu_0)^2 a^4/\lambda^2\right] \tag{13}$$
$$= \exp\left[-\gamma(\mu-\mu_0)^2\right] \text{ (say)},$$

where
$$\gamma = 8\pi^3 N l a^4/\lambda^2. \tag{14}$$

It is now seen that the intensity of the transmitted light is a maximum, being equal to that of the incident light for $\lambda = \lambda_0$. For both smaller and larger wave-lengths, $(\mu-\mu_0)^2$ increases, so that the intensity diminishes. Thus, only a small range of wave-lengths on either side of $\lambda_0$ is transmitted.

It is now interesting to examine how the spectral width of this region of transmission is altered by other factors. As an approximate value of the width, one may take it as equal to the range of wave-lengths, within which the intensity falls to a definite fraction (say $1/e$) of the maximum intensity. Then, $I_l/I_0 = e^{-1}$, so that the width is given by the range of wave-lengths for which

$$\gamma(\mu-\mu_0)^2 < 1, \text{ or } |\mu-\mu_0| < 1/\sqrt{\gamma} \tag{15}$$

Hence, the region of transmission narrows with the increase of $\gamma$, and *vice versa*. The following results immediately follow from this:

(*a*) The region of transmission should sharpen with the increase of the thickness, $l$, of the emulsion and *vice versa*.

(*b*) As the concentration of the dispersed phase is increased, the value of N is increased, so that the spectrum of the transmitted light must become narrower, and *vice versa*.

(*c*) An emulsion containing fine particles must transmit a wider region of the spectrum than one containing coarse droplets, since $a$ is smaller.

(*d*) The more widely different are the relative dispersive powers of the two media, the narrower is the spectral region transmitted. This is so, since the range of values of $\lambda$ within which $(\mu-\mu_0)^2$ becomes equal to $1/\gamma$ is small, if the dispersive powers of the two media are widely different.

All these deductions from the theory have been already verified by Sogani.

In the above discussion, we have not taken the effect of the changes produced in the wave-length. Actually $\gamma$ is inversely proportional to $\lambda^2$, so that it is larger for smaller wave-lengths and *vice versa*. This results in a reduction of the range of wave-lengths transmitted on the shorter wave-length side of $\lambda_0$, and in an increase on the longer wave-length side.

From (13), the value of the attenuation factor is

$$\exp\left[-8\pi^3 N l (\mu-\mu_0)^2 a^4/\lambda^2\right].$$

Now, if one assumes a close-packing of the droplets of the dispersed phase, then the total volume occupied by them is 74 per cent. of the total volume, so that

$$N = \cdot 74/(4\pi a^3/3) = 2\cdot 22/4\pi a^3.$$

If the packing is not so close, one can introduce a coefficient $\sigma$, called the "coefficient of packing", which is equal to the ratio of the actual volume occupied by the dispersed phase to the volume it would occupy in close packing. Then, $N = 2\cdot 22\sigma/4\pi a^3$, so that attenuation factor becomes

$$\exp\left[-4\cdot 44\pi^2\sigma l\,(\mu-\mu_0)^2\,a/\lambda^2\right] = \exp\left[-22\sigma\cdot\frac{(\mu-\mu_0)^2}{\lambda^2}\,ld\right] \qquad (16)$$

where $d$ is the diameter of the droplet.

Sogani has performed some experiments to determine the attenuation coefficient, and found that

$$I_l = I_0 \exp\left[-B\,(\mu-\mu_0)^2\,ld/\lambda^2\right]$$

which is of the same form as the one derived by us. However, for a close-packed emulsion, he found the value $B = 9$, which is much less than the theoretical value derived above. The discrepancy can partly be explained as due to the scattered light also entering the photometer, as suspected by Sogani himself, and also as due to the emulsion not being homogeneous but containing smaller droplets, both of which tend to decrease the attenuation coefficient. But it must also be noted that we have based our theory on the assumption of a random distribution of the particles, with no particular phase correlation. Such a state of affairs cannot be expected to occur in a close-packed emulsion, where one should expect the radiation from the next neighbours at least to be coherent.

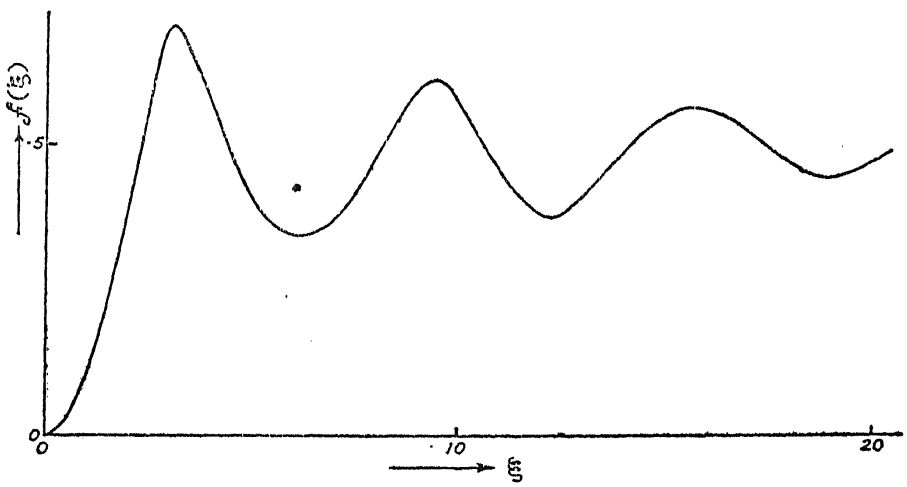


FIG. 1. Graph of $f(\xi)$ against $\xi$

As already remarked, the interesting phenomena exhibited by chromatic emulsions occur only for small values of $\xi$. An evaluation of $\gamma$ for particle

sizes of about 0·003 cm., and a thickness 3 cm. of the emulsion shows that the range of values of $\xi$ within which the light is transmitted is $|\xi| < 0{\cdot}1$. Within this range, $C_1'$ is evidently equal to $\xi^2/8$, the other terms being extremely small. However, it is interesting to examine what happens for much larger values of $\xi$. For these values, the full expression for $C_1'$ can be evaluated from (6) which gives a curve of the type shown in Fig. 1. This curve is the same as Fig. 2 of (1943 *a*), and $f(\xi)$ is the same as the present $C_1'$. From the figure, it is easily seen that the range of phenomena shown by chromatic emulsions lies within the initial portion of the curve, where $C_1'$ increases with $\xi$. But, when $\xi > 3$, $f(\xi)$ must actually diminish with increase of $\xi$, so that in this range, the attenuation coefficient must diminish, and the transmission must increase with increase of $\xi$. This phenomenon would appear worth looking for.

Incidentally, it may be noted that, in the study of transmission of light through water droplets, one is interested in the oscillating portion of the curve, while in the present case it is the limiting portion of the curve near $\xi = 0$ that is important.

### 3. *Refractive Index of the Emulsion*

In the last section, we have shown that the refractive index, $n$, of the emulsion is given by

$$n - \mu_0 = N\lambda a^2 S' \tag{9}$$

$S'$ can also be expanded in powers of $\xi$ as

$$S' = \xi/3 - \xi^3/30 + \cdots \tag{17}$$

and for small values of $\xi$, which only occur in the region of transmission, $S' = \xi/3 = \frac{4\pi}{3}(\mu - \mu_0) \cdot \frac{a}{\lambda}$.

Hence,
$$n - \mu_0 = \frac{4}{3}\pi a^3 \cdot N(\mu - \mu_0).$$

Now, $4\pi a^3 N/3$ is the total volume of the disperse phase, call it $V_0$, contained in unit volume of the emulsion, so that

$$n - \mu_0 = V(\mu - \mu_0), \text{ or}$$

$$n = V\mu + (1 - V)\mu_0 = V\mu + V_0\mu_0, \tag{18}$$

where $V_0$ is the volume of the continuous phase. This shows that the refractive index of the emulsion is the same as that of an ordinary mixture of the two liquids. This is not surprising since, when the difference in the refractive indices of the two media is small, the actual nature of the boundary is unimportant, and the optical behaviour of the emulsion is, as if the two liquids were miscible.

Differentiating (18) with respect to $\lambda$ one gets

$$dn/d\lambda = V d\mu/d\mu + V_0 d\mu_0/d\lambda \tag{19}$$

which gives the relation between the dispersion of the emulsion and those of the two components.

### 4. *Intensity of the Diffracted Light*

The amplitude of the diffracted light is given by expressions (1) to (4), where the integrals in the expression for $X_1$ have to be integrated. Denoting the integrals by C and S, our aim is to integrate them for finite values of $\eta$, so as to be applicable for the case when $\xi$ is small. The method adopted in (1943 *b*) is not suitable in this case, and a new method is therefore used for the purpose.

Expand cos ($\xi$ cos $\theta$) and sin ($\xi$ cos $\theta$) in powers of ($\xi$ cos $\theta$) as

$$\cos(\xi \cos\theta) = 1 - \frac{\xi^2}{(2)!}\cos^2\theta + \frac{\xi^4}{(4)!}\cos^4\theta - \cdots + (-1)^p \frac{\xi^{2p}}{(2p)!}\cos^{2p}\theta + \cdots$$

$$\sin(\xi \cos\theta) = \frac{\xi}{(1)!}\cos\theta - \frac{\xi^3}{(3)!}\cos^3\theta + \cdots + (-1)^p \frac{\xi^{2p+1}}{(2p+1)!}\cos^{2p+1}\theta + \cdots$$

Then,

$$C = \sum_{p=0}^{p=\infty} (-1)^p \frac{\xi^{2p}}{(2p)!} \int_0^{\pi/2} J_0(\eta \sin\theta) \sin\theta \cos^{2p+1}\theta \, d\theta.$$

The integral in each one of the terms of the above series can be integrated, since it is of the standard form of Sonine's first integral, *viz.*,

$$\int_0^{\pi/2} J_\mu(z \sin\theta) \sin^{\mu+1}\theta \cos^{2\nu+1}\theta \, d\theta = \frac{2^\nu \, \Gamma(\nu+1)}{z^{\nu+1}} \cdot J_{\mu+\nu+1}(z)$$

with $\mu = 0$ and $\nu = p$. Hence,

$$C = \sum_{p=0}^{p=\infty} (-1)^p \frac{\xi^{2p}}{(2p)!} \frac{2^p (p)!}{\eta^{p+1}} J_{p+1}(\eta). \tag{20}$$

Similarly, on expanding sin ($\xi$ cos $\theta$), S becomes

$$S = \sum_{p=0}^{\infty} (-1)^p \frac{\xi^{2p+1}}{(2p+1)!} \int_0^{\pi/2} J_0(\eta \sin\theta) \sin\theta \cos^{2p+2}\theta \, d\theta.$$

Here also, each one of the integrals is of the form of Sonine's integral with $\mu = 0$ and $\nu = p + 1/2$, so that

$$S = \sum_{p=0}^{\infty} (-1)^p \frac{\xi^{2p+1}}{(2p+1)!} \frac{2^{p+1/2} \, \Gamma(p+3/2)}{\eta^{p+3/2}} J_{p+3/2}(\eta) \tag{21}$$

On simplifying, and making the substitution $F_\mu(z) = J_\mu(z)/z^\mu$, we get

$$C = \sum_{p=0}^{\infty} (-1)^p \frac{2^p (p)!}{(2p)!} \xi^{2p} F_{p+1}(\eta) \tag{22}$$

$$S = \sqrt{\frac{\pi}{2}}\, \xi \left[ \sum_{p=0}^{\infty} (-1)^p \frac{1}{2^p (p)!} \xi^{2p} F_{p+3/2}(\eta) \right]. \tag{23}$$

The amplitude X is therefore

$$X = K \sin \chi \, [C - F_1(\eta)] - KS \cos \chi. \tag{24}$$

But, $C - F_1(\eta)$ is easily seen to be $= \sum_{p=1}^{\infty} (-1)^p \frac{2^p (p)!}{(2p)!} \xi^{2p} F_{p+1}(\eta)$ which may be represented by $-C_1$. Then,

$$X = -KC_1 \sin \chi - KS \cos \chi. \tag{25}$$

We have thus developed X as a power series in $\xi$, which is convergent for all values of $\xi$ and $\eta$ (*vide* Appendix I). However, the expansion is useful only for small values of $\xi$, for which $C_1$ and S are rapidly convergent, so that the first one or two terms alone need be taken into account. Incidentally, it may be noted that on putting $\eta = 0$, $C_1$ and S become

$$C_1' = \xi^2/8 - \xi^4/144 + \ldots.$$
$$S' = \xi/3 - \xi^3/30 + \ldots.$$

which are identical with the values obtained by direct integration.

The intensity of the light diffracted by a single droplet is, from (25),

$$I = K^2 (C_1^2 + S^2).$$

Substituting for $C_1$ and S, and neglecting terms in $\xi^4$ and higher powers of $\xi$, this becomes*

$$I = \frac{\pi}{2} K^2 \xi^2 F_{3/2}^2(\eta). \tag{26}$$

The above expression relates to the intensity scattered by a single droplet in the direction $\phi$. But the light reaching it, and the light diffracted by it have both to travel through the emulsion, and the total path will not differ from the thickness $l$ of the emulsion, if $\phi$ is not large. Hence, the actual light coming out of the emulsion from one droplet is $I \exp(-\pi a^2 N l \xi^2/2)$. Also, if we take the droplets as distributed at random, then, the intensity of light diffracted out of unit volume of the emulsion in the direction $\phi$ is

$$I_\phi = N \frac{\pi}{2} K^2 \xi^2 F^2_{3/2}(\eta) \exp[-\pi a^2 N l \xi^2/2]. \tag{27}$$

* This expression can also be derived by introducing the approximations directly in equations (1) to (3) as shown in Appendix II.

This is the general expression for the intensity of the diffracted light. In the next two sections, we shall discuss its nature in detail.

### 5. *Light Diffracted by Uniform Emulsions*

In a uniform emulsion, the droplets of the dispersed phase will all be of the same size, so that $a$ is a constant, and the only variable to be considered are the wavelength $\lambda$ and the angle of scattering $\phi$.

(*a*) *Variation of intensity with angle of diffraction* $\phi$

If we use monochromatic light, then $\lambda$ is a constant, so that $\xi$ is fixed. Hence, $I_\phi$ is proportional to $F_{3/2}^2(\eta)$. This quantity is plotted in Fig. 2, from which it will be seen that the intensity is zero for $\eta = 4\cdot5,\ 7\cdot75, \ldots$ and is a maximum for $\eta = 0,\ 6\cdot0,\ 9\cdot1, \ldots$ Since $\eta = 2\pi\,(a/\lambda)\sin\phi$, alternate bright and dark rings must be visible in the diffraction pattern

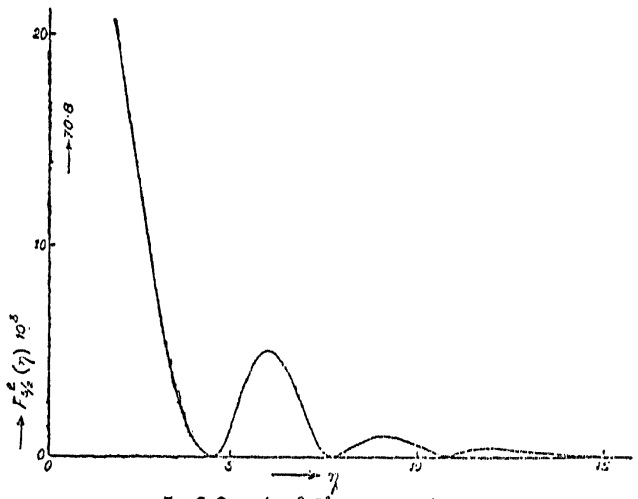


Fig. 2. Graph of $F^2_{3/2}(\eta)$ against $\eta$.

corresponding to the above values of $\eta$. The relative intensities of the first three bright rings are given by $I_1 : I_2 : I_3 = 5 : 1 : 0\cdot25$. For $a = 2 \times 10^{-3}$ and $\lambda = 6 \times 10^{-5}$, the angle of diffraction for the first bright ring comes out to be $\phi = 0\cdot028^c$. In fact, Sogani, using a homogenized emulsion, found a value $0\cdot025^c$ for the angular radius of the first ring. The agreement of this with the theoretical value is not to be stressed, for the type of pattern observed by Sogani had a minimum of intensity at the centre, while our theory requires a maximum. The effect observed by Sogani is similar to the diffraction of X-rays by liquids; on account of the fact that the droplets in a homogenized emulsion have a quasi-regularity in arrangement, interference effects arise in addition to the diffraction by individual droplets.

(*b*) *Variation of the Intensity with Wavelength*

From the expression (27), it is seen that the intensity $I_\phi \propto \xi^2 e^{-\beta\xi^2} F_{3/2}^2(\eta)$ where $\beta = \pi a^2 Nl/2$. Since $F_{3/2}^2(\eta)$ gives only a variation with angle, the

intensity of the ring system as a whole can be said to be proportional to $\xi^2 e^{-\beta\xi^2}$. This quantity is equal to zero at $\xi = 0$, increases with $\xi$ up to $\xi^2 = 1/\beta$ and then decreases again. Hence, no ring system must be visible at $\lambda = \lambda_0$, for which $\xi = 0$. On either side, the rings must appear, and at first increase in intensity as the wavelength is removed more and more from $\lambda_0$, until it reaches a maximum. Thereafter, the intensity must diminish again until, when $\lambda$ is far removed from $\lambda_0$, no rings must be visible. All these have been experimentally verified by Sogani.

If now, white light is used, and the spectrum of the light diffracted at an angle is observed, then the intensity will vary as $\xi^2 e^{-\beta\xi^2}$, being zero for $\lambda = \lambda_0$, and being a maximum for the values of $\lambda$ for which

$$\xi^2 = 1/\beta \text{ or } (\mu - \mu_0)^2 = 1/\gamma. \tag{28}$$

The function $\xi^2 e^{-\beta\xi^2}$ has been plotted in Fig. 3 against $\xi$ as ordinate, for the value 2 of $\beta$. This shows the nature of the intensity distribution in the spectrum of the diffracted light.

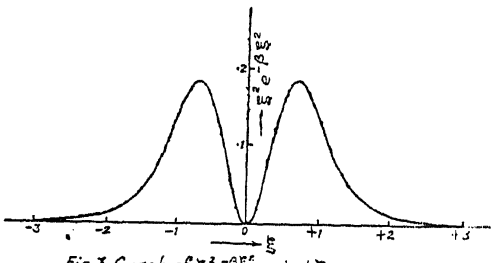


Fig. 3. Graph of $\xi^2 e^{-\beta\xi^2}$ against $\xi$

(c) *Variation of the Size of the Rings with the Wavelength*

Since $\eta = 2\pi a \sin(\phi)/\lambda$, it is evident that as $\lambda$ increases, the size of the rings must increase. Thus, the width of the ring system must *continuously* increase with the wavelength, although its intensity is a minimum at $\lambda_0$, and increases on either side. This fact has also been noted by Sogani.

Another interesting fact, also noted by Sogani, is that "the size of the rings....is, strangely enough, uninfluenced by the thickness of the emulsion". This, however, is a natural conclusion from our theory, for N and $l$ occur only in the exponential in (27), and do not occur in the function $F_{3/2}^2(\eta)$, so that, for the same reason as explained in Section (4) of (1943 *b*), the size of the rings must be uninfluenced by the thickness ($l$) or the concentration (N) of the emulsion.

## 6. *Phenomena with Non-Uniform Emulsions*

In a non-uniform emulsion, all the droplets are not of the same size, so that the phenomena are a little complicated. In the preceding section,

it has been shown that the intensity diffracted in a direction $\phi$ is $\frac{\pi}{2} K^2 \xi^2 F_{3/2}{}^2(\eta)$. Hence, the intensity is directly proportional to the sixth power of the radius of the droplet. But the width of the bright central portion is determined by the value of $\eta$, which must be less than $4 \cdot 5$, *i.e.*, the range of angles covered by the central bright portion is given by $\sin \phi < 4 \cdot 5\, \lambda / 2\pi a$. Thus, the width of this portion diminishes with increase of the size of the particle. Hence, it is clear that, for any angle of diffraction $\phi_1$, the maximum intensity will be due to particles of a certain radius $a_1$ (say), those having a larger or a smaller radius, giving only a less intensity. This can be proved as follows:

If the angle of diffraction is $\phi_1$, then $\eta = 2\pi \sin(\phi_1)\, a/\lambda$, and $\xi = 4\pi(\mu - \mu_0)\, a/\lambda$, so that for a particular wavelength, $\xi$ is a constant multiple of $\eta$. Also, K is a constant multiple of $\xi$, so that

$$I_\phi \propto \eta^6 J_{3/2}{}^2(\eta)/\eta^3 \propto \eta^3 J_{3/2}{}^2(\eta) \tag{29}$$

Hence $I_\phi$ is a maximum for that value, $\eta_0$, of $\eta$ for which $\eta^3 J_{3/2}{}^2(\eta)$ is a maximum. The radius of the droplet corresponding to this is given by $2\pi \sin(\phi_1)\, a_1/\lambda = \eta_0$, or

$$a_1 = \lambda \eta_0 / 2\pi \sin \phi_1. \tag{30}$$

Thus, for every angle $\phi_1$, the predominant portion of the diffracted light is due to particles of radius $a_1$ given by (30), which is smaller for larger angles.

In Section 5, we have shown that the spectrum of the diffraction light always consists of a minimum at $\lambda = \lambda_0$, and two maxima on either side corresponding to

$$(\mu - \mu_0)^2_{max.} = 1/\gamma. \tag{28}$$

But $\gamma$ is a function of $a_1$, so that it varies with the angle of diffraction in the present case. Since $\gamma = 8\pi^3 N l \cdot a^4/\lambda^2$, for an angle of diffraction $\phi_1$,

$$\gamma = \frac{8\pi^3 N l\, \lambda^4 \eta_0{}^4}{16\, \pi^4 \sin^4 \phi_1\, \lambda^2} = \frac{N l\, \eta_0{}^4\, \lambda^2}{2\pi \sin^4 \phi_1}. \tag{31}$$

Thus, other quantities being the same,

$$(\mu - \mu_0)_{max.} \propto \sin^2 \phi_1. \tag{32}$$

If, in the immediate neighbourhood of $\lambda_0$, we assume a direct proportionality between $(\mu - \mu_0)$ and $(\lambda - \lambda_0)$, then it at once follows that the spectral range between the two maxima, *viz.*,

$$2(\lambda - \lambda_0)_{max.} \propto 2(\mu - \mu_0)_{max.} \propto \sin^2 \phi_1, \tag{33}$$

Thus, as the angle of diffraction $\phi$ is increased, the central dark portion in the spectrum of the diffracted light must widen, and correspondingly the bright portions also must broaden. This result from the theory is remarkably confirmed by Sethi's observations, where a progressive broadening of

the spectrum was found as the direction of observation was taken further away from the incident direction.

In the discussion given above, we have neglected the effect of the variation of $\lambda$. On introducing this also, $\gamma \propto \lambda^2/\sin^4 \phi_1$, so that

$$(\mu - \mu_0)_{max.} \propto \sin^2 \phi_1/\lambda. \tag{34}$$

Thus, the quantity $(\mu - \mu_0)_{max.}$ varies continuously with $\lambda$, being greater for smaller wavelengths. Hence, at large angles, where an appreciable portion of the spectrum is transmitted, there must be a further broadening on the shorter wavelength side, and an opposite effect on the longer wavelength side. So also, the actual intensity is proportional to $\lambda^{-4}$, which enormously increases the intensity for smaller wavelengths. For both these reasons, the spectrum must show an asymmetry, greater intensity being concentrated on the shorter wavelength side of the transmission band.

Another assumption made in the above discussion is that droplets of all sizes are present in the emulsion. Actually, there must be an upper and a lower limit to the size of the droplets. This fact limits the minimum and the maximum widths of the spectrum. Between these limits, however, the width must regularly increase with the angle of diffraction.

My sincere thanks are due to Prof. Sir C. V. Raman, for suggesting the problem and for the keen interest that he took in it.

*Summary*

A theory of the optical phenomena exhibited by chromatic emulsions as also those observed in the Christiansen experiment has been worked out, *de novo*, on the basis of the diffraction of light by a sphere immersed in a medium of nearly the same refractive index. Expressions are derived both for the intensity of the transmitted light, and of the light diffracted in other directions. These are discussed in relation to the intensity and the spectral nature of the light and it is shown that the theory can satisfactorily account for the various phenomena observed by Sethi and Sogani.

REFERENCES

1. Christiansen .. *Ann. d. Phys.*, 1884, **23**, 298.
2. Holmes, H. N., and Cameron, D. H. .. *Journ. Am. Chem. Soc.*, 1922, **44**, 71.
3. Ramachandran, G. N. *Proc. Ind. Acad. Sci.* **A**, 1943 *a*, **17**, 171.
4. ———— .. *Ibid.*, 1943 *b*, **17**, 202.
5. Rayleigh, Lord .. *Phil. Mag.*, 1885, **20**, 358.
6. ———— .. *Sci. Papers*, 1899, **IV**, 392.
7. Sethi, N. K. .. *Proc. Ind. Assoc. Cult. Sci.*, 1921, **6**, 124.
8. Sogani, C. M. .. *Phil. Mag.*, 1926, **1**, 321.

## APPENDIX I

*Note on the Convergency of the Series $C_1$ and $S$*

Let $C_1 = \Sigma u_p = \sum_{p=1}^{\infty} (-1)^p \frac{2^p (p)!}{(2p)!} \xi^{2p} \frac{J_{p+1}(\eta)}{\eta^{p+1}}$.

Then $\left|\frac{u_{p+1}}{u_p}\right| = \frac{2(p+1)}{(2p+1)(2p+2)} \cdot \frac{\xi^2}{\eta} \cdot \frac{J_{p+2}(\eta)}{J_{p+1}(\eta)}$.

But, for large values of $p$, $J_{p+2}(\eta)/J_{p+1}(\eta) = \eta/2(p+2)$, so that

$\underset{p\to\infty}{\mathrm{Lt}} \left|\frac{u_{p+1}}{u_p}\right| = \underset{p\to\infty}{\mathrm{Lt}} \frac{\xi^2}{(2p+1)(2p+2)} \cdot \frac{(p+1)}{(p+2)}$ which is $=0$, if $\xi$ is finite.

Hence $\Sigma u_p$ is absolutely convergent.

In the same way, putting

$$S = \Sigma v_p = \sum_{p=0}^{\infty} (-1)^p \frac{1}{2^p (p)!} \xi^{2p+1} \frac{J_{p+3/2}(\eta)}{\eta^{p+3/2}},$$

$$\left|\frac{v_{p+1}}{v_p}\right| = \frac{1}{2p} \cdot \frac{\xi^2}{\eta} \cdot \frac{J_{p+5/2}(\eta)}{J_{p+3/2}(\eta)}.$$

Substituting the value for $J_{p+5/2}(\eta)/J_{p+3/2}(\eta)$ for large values of $p$, namely $\eta/(2p+5)$,

$\underset{p\to\infty}{\mathrm{Lt}} \left|\frac{v_{p+1}}{v_p}\right| = \underset{p\to\infty}{\mathrm{Lt}} \frac{\xi^2}{2p(2p+5)}$, which is also $=0$, if $\xi$ is finite.

Thus, $\Sigma v_n$ is also absolutely convergent.

Hence, both $C_1$ and S are convergent for all values of $\xi$ and $\eta$.

## APPENDIX II

*A Simple Derivation of the Expression for the Intensity*

As already remarked, one is interested only in small values of $\xi$ in the study of chromatic emulsions, in which case $\cos(\xi \cos\theta) \simeq 1$ and $\sin(\xi\cos\theta) \simeq (\xi\cos\theta)$. Substituting these in (1) to (3),

$$X = K \sin\chi \left[\int_0^{\pi/2} J_1(\eta\sin\theta)\sin\theta\cos\theta\, d\theta - J_1(\eta)/\eta\right]$$

$$- K\cos\chi \int_0^{\pi/2} J_0(\eta\sin\theta)(\xi\cos\theta)\sin\theta\cos\theta\, d\theta$$

The quantity within the square brackets vanishes, and using Sonine's integral with $\mu = 0$ and $\nu = 1/2$, the second term becomes

$$-\sqrt{\frac{\pi}{2}}\, K\, \xi\, J_{3/2}(\eta)/\eta^{3/2},$$

Hence, the intensity is

$$I = \frac{\pi}{2} K^2 \xi^2 J_{3/2}^2(\eta)/\eta^3,$$

which is identical with expression (26).