

The Royal Society  
Burlington House  
London W. 1

113

21 May 1947

Sir,

The paper communicated by you to the  
Royal Society, entitled

Light-scattering in homogeneous media  
regarded as reflection from appropriate  
thermal elastic waves

A. 59

has been safely received.

Yours faithfully,

*D. C. Martin*

*Assistant Secretary*

IN SO FAR AS THE PAPER COMMUNICATED BY YOU HAS BEEN  
CARRIED OUT IN OR ON BEHALF OF A GOVERNMENT RESEARCH  
ESTABLISHMENT, WILL YOU KINDLY STATE IN WRITING WHETHER  
THE AGREEMENT OF THE DEPARTMENT CONCERNED HAS BEEN  
OBTAINED TO PUBLICATION OF THE PAPER.

*John D. Martin*  
*D. C. Martin*

Assistant Secretary

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*Assistant Secretary*

Received 21 May 1947

MS. 1

Figures -

Plates (Photographs, etc.)

Society  
Institution House  
London W.1

8 August 1947.

Dear Sir,

I have to inform you that the paper No. A.59  
by A.B. Bhatia and yourself

which you communicated to the Society has been accepted for  
publication in the Proceedings/~~Transactions~~

Galley proofs will be sent for your approval in due  
course. If you require the proofs to be read by any person other  
than yourself, please let me know the name and address to which  
they should be sent.

Yours faithfully,

D. C. Martin

Assistant Secretary

Recd 6<sup>10</sup>/<sub>17</sub> +  
replied via same  
day <sup>STP</sup> requesting  
proof be sent to  
Bhatia at Bristol  
use.

Blanket

L.P. 10/10/47 oc

Registered



Sir, Kariamanikkam Krishnan, F.R.S.

*Director*

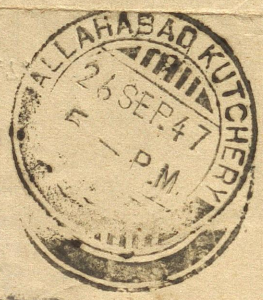
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DELHI



Published in Proc. Roy. Soc. A. Feb. 1948

Light-scattering in homogeneous media regarded as reflection from appropriate thermal elastic waves

by

Sir K.S. Krishnan, F.R.S., and A.B. Bhatia, D.Phil.,  
University of Allahabad.

A B S T R A C T

*stationary!*

Attributing the scattering of light in a homogeneous transparent liquid to the local fluctuations in density, and the latter to the superposition of the thermal elastic waves of different wave-length maintained in the enclosure, one may, following Einstein, evaluate the scattering coefficient of the liquid along any given direction. The expression for the scattering coefficient involves a triple infinite series, which Einstein evaluates by suitably replacing it by a triple integral. The series, however, can be directly summed, and the contributions from ~~neighbouring~~ <sup>different</sup> progressive waves to scattering studied in detail. This method brings out prominently the appropriateness of regarding scattering as regular Bragg reflections from suitable elastic waves; and also reveals <sup>some</sup> many interesting features which are missed when the summation is replaced by integration.

The intensities of the Brillouin components, are calculated on this basis, both in a fluid medium and in a crystal; and in the latter case the expression for the over-all intensity of the Brillouin components is shown to be identical with the well-known expression of Waller in X-ray scattering.

# 1. EINSTEIN'S TREATMENT OF LIGHT-SCATTERING IN A LIQUID

The intensity of light scattered by a homogeneous liquid is calculated by Einstein (1910) in the following manner. Consider for simplicity a monatomic liquid contained in a large cube of edge-length  $L$ ,  $0 < x < L$ ,  $0 < y < L$ ,  $0 < z < L$ . The local fluctuations in the density of the liquid, due to thermal agitation, which ultimately produce the scattering, may be expressed in terms of the stationary elastic waves maintained in the cube ;

$$D = D_0 + \Delta$$

$$\Delta = \sum_{\rho} \sum_{\sigma} \sum_{\tau} B_{\rho\sigma\tau} \sin \frac{2\pi\rho x}{2L} \cdot \sin \frac{2\pi\sigma y}{2L} \cdot \sin \frac{2\pi\tau z}{2L} e^{i(2\pi N_{\rho\sigma\tau} t - \theta_{\rho\sigma\tau})}, \dots (1)$$

where  $\Delta$  is the fluctuation in density at any point  $xyz$  from the mean value  $D_0$ , and  $\rho\sigma\tau$  are positive integers which define as usual the stationary elastic waves; a stationary wave defined by given  $\rho\sigma\tau$  will consist in general of eight progressive waves whose wave-length  $\Lambda_{\rho\sigma\tau}$  is equal to  $2L / (\rho^2 + \sigma^2 + \tau^2)^{1/2}$ , and the direction-cosines of whose wave-normals (one in each octant of coordinate space) are given by the eight combinations of  $(\pm\rho, \pm\sigma, \pm\tau) / (\rho^2 + \sigma^2 + \tau^2)^{1/2}$ .

$$N_{\rho\sigma\tau} = V_{\rho\sigma\tau} / \Lambda_{\rho\sigma\tau}, \dots (2)$$

where  $N_{\rho\sigma\tau}$  is the frequency, and  $V_{\rho\sigma\tau}$  is the phase velocity, of elastic waves of wave-length  $\Lambda_{\rho\sigma\tau}$ .

Consider a small cube of edge-length  $l$  of the liquid, with its edges parallel to those of the large cube. The fraction of the incident light scattered by this element of volume  $l^3$ , per unit solid angle, along a direction making an angle  $\varphi$  with the incident direction, will then be given

by

$$S_{\varphi} l^3 = \frac{l^6}{64D_0^2} n^2 \sigma_{\varphi} \sum_{\rho} \sum_{\sigma} \sum_{\tau} \overline{B_{\rho\sigma\tau}^2} \frac{\sin^2 \xi}{\xi^2} \cdot \frac{\sin^2 \eta}{\eta^2} \cdot \frac{\sin^2 \zeta}{\zeta^2}, \dots (3)$$

where  $S_{\varphi}$  is the scattering coefficient per unit volume of the liquid per unit solid angle along the direction considered, and  $\sigma_{\varphi}$  is the corresponding coefficient for an individual (not isolated) atom in the liquid,  $n$  is the number of atoms per unit volume,  $\overline{B_{\rho\sigma\tau}^2}$  is the time-average of  $B_{\rho\sigma\tau}^2$ , and

$$\left. \begin{aligned} \xi &= \frac{\pi}{2} \cdot \frac{l}{L} (\rho - \rho_0) \\ \eta &= \frac{\pi}{2} \cdot \frac{l}{L} (\sigma - \sigma_0) \\ \zeta &= \frac{\pi}{2} \cdot \frac{l}{L} (\tau - \tau_0) \end{aligned} \right\} \dots (4)$$

where  $\rho_0, \sigma_0, \tau_0$  are positive numbers, not necessarily integers, which analogously to  $\rho, \sigma, \tau$  define certain wave-normal directions, and a wave-length, and are such that

- (1) one of the four pairs of wave-normals defined by them is along the bisector of the exterior angle  $\pi - \varphi$  between the directions of incidence and of observation,
- (2) the wave-length  $\Lambda_0 = 2L / (\rho_0^2 + \sigma_0^2 + \tau_0^2)^{\frac{1}{2}}$  defined by them satisfies the Bragg condition for reflecting the incident light-waves along the direction of observation, namely

$$2 \Lambda_0 \sin \frac{\varphi}{2} = \lambda, \dots (5)$$

where  $\lambda$  is the wave-length of the incident light.

In other words, if there were elastic waves in the liquid of the above wave-length, and progressing along the above bisector in either direction, the coefficient of reflection of the incident light-waves from these elastic waves would be a maximum in the direction selected for observation of

scattering.

In (3) we have expressed the scattering coefficient  $S_{\varphi}$  of the liquid along any given direction  $\varphi$  in terms of the scattering coefficient of the atom, i.e., the atomic cross-section for scattering, along the same direction, namely  $\sigma_{\varphi}$ . The  $\sigma_{\varphi}$  introduced here is analogous to the well-known atom form factor for intensity in X-ray scattering.  $\sigma_{\varphi}$  will be proportional to  $M^2$  where  $M$  is the dipole moment induced in the atom per unit "field in the medium", and since the actual field acting on the atom which produces the dipole moment  $M$  is not merely the field in the medium but includes the polarization field due to surrounding atoms,  $\sigma_{\varphi}$  will not be an atomic constant, but will depend also on the density of packing of the atoms in the medium.

We may mention here that in Einstein's derivation,  $S_{\varphi}$  is expressed in terms of the local fluctuations in the refractivity of the liquid accompanying the fluctuations in density. We shall discuss the value of  $\sigma_{\varphi}$  in the liquid in relation to the density of packing of the atoms, and the refractivity of the liquid, in a later section of the paper (See Section 6).

We should, however, emphasise here a fundamental assumption that underlies the derivation of relation (3). Consider a small element of volume  $v$  in the liquid containing on an average  $N$  atoms, the linear dimensions and the disposition of the volume element being such that the scattered radiations from the different parts of the element reach the observer in practically the same phase. If  $\overline{\Delta N^2}$  is the mean square of the fluctuation in the number of atoms in the volume element from its average value  $N$ , the assumption referred to is that the contribution from this volume element to the scattering by the liquid is given by

$$S_{\varphi} v = \overline{\Delta N^2} \sigma_{\varphi}. \quad \dots (6)$$

We shall discuss the validity and the implications of this assumption when we take up in Section 6 below the question of the relation between the atomic scattering coefficient in the liquid, and the density and the refractivity of the liquid.

## 2. EVALUATION OF THE SERIES APPEARING IN (3) BY INTEGRATION

Coming back to expression (3) for  $S_{\varphi}$ , since the wave-length of light is long in comparison with the inter-atomic distance, the values of  $\rho_0 \sigma_0 \tau_0$ , even for the backward direction of observation, will be much smaller than the maximum values of  $\rho \sigma \tau$  for the elastic waves, and since the significant values of  $\sin^2 \xi / \xi^2$ ,  $\sin^2 \eta / \eta^2$ , and  $\sin^2 \zeta / \zeta^2$  are confined to small values of  $\xi$ ,  $\eta$  and  $\zeta$  respectively, the summations in (3) may be taken to extend over all permitted (discrete) values of  $\xi$  or  $\eta$  or  $\zeta$  as the case may be, from  $-\infty$  to  $+\infty$ .

Now  $\rho \sigma \tau$  vary in steps of unity, and for given directions of incidence and observation, i.e. for given  $\rho_0 \sigma_0 \tau_0$ , the corresponding steps in the variation of  $\xi \eta \zeta$  will be  $\pi/2 \cdot \ell/L$ , and can be made infinitesimally small by making  $\ell$  sufficiently small in comparison with  $L$ . Doing so, and regarding  $\overline{B_{\rho\sigma\tau}^2}$  as independent of  $\rho \sigma \tau$  — which will be practically the case at room temperature, which is high enough for the thermal energies of the long elastic waves that are involved here to be nearly proportional to the absolute temperature — we obtain

$$\begin{aligned} \sum \sum \sum \overline{B_{\rho\sigma\tau}^2} \frac{\sin^2 \xi}{\xi^2} \cdot \frac{\sin^2 \eta}{\eta^2} \cdot \frac{\sin^2 \zeta}{\zeta^2} &= \overline{B^2} \left( \frac{2}{\pi} \cdot \frac{L}{\ell} \right)^3 \int_{-\infty}^{+\infty} \frac{\sin^2 \xi}{\xi^2} \cdot \frac{\sin^2 \eta}{\eta^2} \cdot \frac{\sin^2 \zeta}{\zeta^2} d\xi d\eta d\zeta \\ &= \overline{B^2} \left( \frac{2L}{\ell} \right)^3, \quad \dots (7) \end{aligned}$$

from which we obtain

b<sup>2</sup>

$$S_{\varphi} = n^2 \sigma_{\varphi} \cdot \frac{\overline{B^2} L^3}{8 D_0^2}, \quad \dots (8)$$

At the high temperatures that we are considering

$$\frac{\overline{B^2} L^3}{8 D_0^2} = k T \beta, \quad \dots (9)$$

where  $\beta$  is the isothermal compressibility of the liquid, and  $k$  is the Boltzmann constant, and we obtain for the scattering coefficient per unit volume, per unit solid angle, along the direction considered

$$S_{\varphi} = n^2 \sigma_{\varphi} \cdot k T \beta. \quad \dots (10)$$

For a gas obeying Boyle's law,  $n k T \beta = 1$ , and  $S_{\varphi}$  becomes equal to  $n \sigma_{\varphi}$ , as should be expected since the scattered radiations even from neighbouring atoms will then be of random phases. In the other extreme case where all the atoms scatter in the same phase,  $S_{\varphi}$  will evidently be equal to  $n^2 \sigma_{\varphi}$ .

### 3. DIRECT SUMMATION OF THE SERIES

We wish to point out here that the series appearing on the left hand side of (7) may be summed up directly, and to draw attention to certain interesting features in scattering that are revealed by the summation, and are missed when it is replaced by integration in the manner described above. What is required is the sum of the values of  $\sin^2 \xi / \xi^2$ , etc., at equal intervals  $\alpha = \pi/2 \cdot l/L$ , and it can be shown that

$$\sum_{n=-\infty}^{+\infty} \frac{\sin^2 (n\alpha + \theta)}{(n\alpha + \theta)^2} = \frac{\pi}{\alpha} \quad \dots (11)$$

where  $n$  is an integer,  $0 < \alpha \leq \pi$ , and  $\theta$  is a constant. ( $\theta$  in our problem is equal to  $\pi/2 \cdot l/L$  times the fractional part of  $\rho_0$  or  $\sigma_0$  or  $\tau_0$ , as the case may

be). The proof is as follows.†

Consider the known series\*

$$\sum_{n=-\infty}^{+\infty} \frac{\sin\{(n+\beta)\gamma\}}{n+\beta} = \pi, \quad \dots (12)$$

in which  $\beta$  is a constant,  $n$  an integer, and  $0 < \gamma < 2\pi$ . The series can be integrated term by term with respect to  $\gamma$  in any closed interval  $(\gamma, \delta)$ , where  $0 < \gamma < \delta < 2\pi$ , since it is uniformly convergent in this interval. We then obtain

$$\sum_{n=-\infty}^{+\infty} \frac{\cos\{(n+\beta)\gamma\}}{(n+\beta)^2} - \sum_{n=-\infty}^{+\infty} \frac{\cos\{(n+\beta)\delta\}}{(n+\beta)^2} = \pi(\delta - \gamma). \quad \dots (13)$$

Keeping  $\delta$  constant and making  $\gamma \rightarrow 0$ , it is readily seen that (13) reduces to

$$\sum_{n=-\infty}^{+\infty} \frac{1 - \cos\{(n+\beta)\delta\}}{(n+\beta)^2} = \pi\delta,$$

since the first series on the left hand side of (13) is uniformly convergent, and therefore represents a continuous function of  $\gamma$ . Putting now  $\delta = 2\alpha$ , and  $\alpha\beta = \theta$ , and dividing both sides by  $2\alpha^2$  ( $\alpha \neq 0$ ), we obtain, for  $0 < \alpha < \pi$ ,

$$\sum_{n=-\infty}^{+\infty} \frac{\sin^2(n\alpha + \theta)}{(n\alpha + \theta)^2} = \frac{\pi}{\alpha}. \quad \dots (11)$$

This can be seen to be true for  $\alpha = \pi$  also, and hence (11) holds over the interval  $0 < \alpha \leq \pi$ .†

† Proof rewritten October 28, 1947.

\* See for example I. Bromwich, "An Introduction to the Theory of Infinite Series", Macmillan, 1931, p.371, ex. 5.

† We are thankful to Professor Norbert Wiener for the following elegant alternative proof of (11). Let

$$\text{Let } g(v) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} f(u) e^{iuv} du$$

be the Fourier transform of the function

$$f(u) = \frac{\sin^2(u + \theta)}{(u + \theta)^2} = \frac{\sin^2 w}{w^2}, \text{ say.}$$

Then

$$\begin{aligned} g(v) &= \frac{e^{-iv\theta}}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} \frac{\sin^2 w}{w^2} e^{i w v} dw \\ &= \frac{e^{-iv\theta}}{\sqrt{2\pi}} \pi \left[ 1 - \frac{|v|}{2} \right], \neq 0 \text{ when } -2 < v < +2. \end{aligned}$$

Now according to Poisson's formula,

*this footnote is continued at the foot of next page.*

*Keep the original proof see next p*

be ). The proof is as follows:

Eqn (12) holds only if  $0 < y < 2\pi$ .

Consider the well-known series\*  

$$\sum_{n=1}^{\infty} \left[ \frac{\sin\{(n+\beta)y\}}{n+\beta} + \frac{\sin\{-n+\beta)y\}}{-n+\beta} \right] = -\frac{\sin \beta y}{\beta} + \pi, \quad 0 < y < 2\pi \quad (12)$$

in which  $\beta$  is a constant, and  $n$  an integer. Confining ourselves to the interval  $0 \leq y < 2\pi$ , this series in (12) can be integrated term by term (i.e., the sum of the series of integrals thus obtained will be equal to the integral of the sum), since it is uniformly convergent <sup>except</sup> in the neighbourhood of zero, and is bounded <sup>by</sup> convergent over the whole interval including zero. Replacing each sine term in (12) by the product of a sine and <sup>a</sup> cosine term and integrating, we have

$$2 \sum_{n=1}^{\infty} \left[ \frac{\sin^2\{(n+\beta)y/2\}}{(n+\beta)^2} + \frac{\sin^2\{-n+\beta)y/2\}}{(-n+\beta)^2} \right] = \frac{2}{\beta^2} \sin^2 \frac{\beta y}{2} + \pi y \dots (13)$$

Putting  $y/2 = \alpha$ ,  $\alpha\beta = \theta$ , transposing  $\frac{2}{\beta^2} \sin^2 \frac{\beta y}{2}$  to the left hand side, and dividing both sides by  $\alpha^2$ ,  $\alpha \neq 0$ ,

we obtain for  $0 < \alpha < \pi$

$$\sum_{n=-\infty}^{+\infty} \frac{\sin^2(n\alpha + \theta)}{(n\alpha + \theta)^2} = \frac{\pi}{\alpha}$$

This can be readily seen to be true for  $\alpha = \pi$  also, and hence (11) holds over the interval  $0 < \alpha \leq \pi$ . †

\* See for example J. Bromwich, "An Introduction to the Theory of Infinite Series", Macmillan, 1931, p. 371 <sup>ex. 5.</sup>

† We are thankful to Professor Norbert Wiener for the following elegant alternative proof of (11).

Let  $g(v) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} f(u) e^{iuv} du$

be the Fourier transform of the function

$$f(u) = \frac{\sin^2(u + \theta)}{(u + \theta)^2} = \frac{\sin^2 w}{w^2}, \text{ say,}$$

Then

$$g(v) = \frac{e^{-iv\theta}}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} \frac{\sin^2 w}{w^2} e^{i w v} dw$$

$$= \frac{e^{-iv\theta}}{\sqrt{2\pi}} \pi \left[ 1 - \frac{|v-1|}{2} \right], \neq 0 \text{ when } -2 < v < 2.$$

Now according to Poisson's formula,

First note comes on next page

We notice in particular that the sum in (11) is independent of  $\theta$ .

Coming back to the series appearing on the left hand side of (7), we obtain using (11)

$$\sum_{\xi} \sum_{\eta} \sum_{\zeta} \overline{B^2} \frac{\sin^2 \xi}{\xi^2} \cdot \frac{\sin^2 \eta}{\eta^2} \cdot \frac{\sin^2 \zeta}{\zeta^2} = \overline{B^2} \left( \frac{\pi}{\alpha} \right)^3 = \overline{B^2} \left( \frac{2L}{\ell} \right)^3, \dots (14)$$

the same result as obtained in (7) by integration.

Since the series can be summed up, it is not necessary to make  $\ell$  small. One may choose  $\ell$  as large as one likes, even make it equal  $L$ , in which case  $\alpha$  becomes equal to  $\pi/2$ , and still satisfies the condition  $0 < \alpha \leq \pi$ .

Making  $\ell = L$ , one obtains from (3)

$$S_{\varphi} = n^2 \sigma_{\varphi} \cdot \frac{L^3}{64 D_0^2} \overline{B^2} \sum_{\rho} \sum_{\sigma} \sum_{\tau} \dots (15)$$

where

$$\sum_{\rho} = \sum_{n=-\infty}^{+\infty} \frac{\sin^2 \left( \frac{n\pi}{2} + \theta_{\rho} \right)}{\left( \frac{n\pi}{2} + \theta_{\rho} \right)^2}, \text{ etc.}, \dots (16)$$

and  $\theta_{\rho} = \pi/2$  times the fractional part of  $\rho$ , etc.

Each of the three sums appearing in (15) is obviously equal to 2, giving for  $S_{\varphi}$  the same value <sup>(8)</sup> as before.

#### 4. LIGHT-SCATTERING REGARDED AS REFLECTION FROM APPROPRIATE ELASTIC WAVES

Expressions (15) and (16) for  $S_{\varphi}$  give us the following information :

(1) For any given stationary wave (specified by given

$$\sum_{n=-\infty}^{+\infty} f(n\alpha) = \frac{\sqrt{2\pi}}{\alpha} \sum_{n=-\infty}^{+\infty} g\left(\frac{2\pi n}{\alpha}\right),$$

where  $n$  is an integer, and  $\alpha > 0$ . If further, as in our problem,  $\alpha$  is not greater than  $\pi$ , there is only one value of  $n$  for which  $g\left(\frac{2\pi n}{\alpha}\right)$  differs from 0, namely  $n = 0$ ; and  $g(0) = \sqrt{\pi/2}$ , and is independent of  $\theta$ . Hence

$$\sum_{n=-\infty}^{+\infty} f(n\alpha) = \frac{\sqrt{2\pi}}{\alpha} \cdot \sqrt{\frac{\pi}{2}} = \frac{\pi}{\alpha}.$$

$\rho \sigma \tau$  (15) and (16) give the variation of its contribution to  $S_{\varphi}$  with the change in the direction of incidence, or of observation, i.e. with the change in  $\rho_0 \sigma_0 \tau_0$ . The variation is as

$$\sin^2 \xi / \xi^2 \cdot \sin^2 \eta / \eta^2 \cdot \sin^2 \zeta / \zeta^2$$

and  $\xi = 0$  when  $\rho = \rho_0$ , etc., and

$$d\xi/d\rho_0 = d\eta/d\sigma_0 = d\zeta/d\tau_0 = \pi/2.$$

- (2) For any given directions of incidence and of observation, i.e., for any given  $\rho_0 \sigma_0 \tau_0$  (15) and (16) give the contributions to  $S_{\varphi}$  from different stationary waves, i.e. stationary waves of differing  $\rho \sigma \tau$ ; these contributions also are proportional to  $\sin^2 \xi / \xi^2 \cdot \sin^2 \eta / \eta^2 \cdot \sin^2 \zeta / \zeta^2$ ,  $\xi$  being zero when  $\rho = \rho_0$  etc., and a change of  $\rho$  or  $\sigma$  or  $\tau$  by unity, as will occur when we pass from one stationary wave to the adjacent one, corresponding to a change of  $\pi/2$  in  $\xi$ , or  $\eta$ , or  $\zeta$ , respectively.

It can be readily seen that the resolving power of the stationary waves regarded as forming a reflection grating is just half that necessary to resolve, in the Rayleigh sense, the reflections from adjacent stationary waves, by which we mean stationary waves whose  $\rho$  or  $\sigma$  or  $\tau$  values differ by unity. Hence the significant part of the contribution to  $S_{\varphi}$  comes from waves whose  $\rho \sigma \tau$  lie close to  $\rho_0 \sigma_0 \tau_0$ , and practically lie in the range  $\rho_0 \pm 2$ ,  $\sigma_0 \pm 2$ ,  $\tau_0 \pm 2$ . Hence it is not now necessary to assume the independence of  $\overline{B_{\rho\sigma\tau}^2}$  on  $\rho \sigma \tau$ . We choose for  $\overline{B^2}$  the value appropriate to the neighbourhood of  $\rho_0 \sigma_0 \tau_0$ .

On the other hand when the scattering volume is restricted to a small element  $l^3$ ,  $l \ll L$ , the resolving power of the elastic waves inside the element regarded as forming a

reflection grating naturally becomes smaller by a factor  $l/L$ , and hence not only the few elastic waves whose  $\rho \sigma \tau$  values lies in the close neighbourhood of  $\rho_0 \sigma_0 \tau_0$ , but waves corresponding to a much wider range  $-(\rho_0 - 2L/l) < \rho < (\rho_0 + 2L/l)$  etc. — will apparently contribute to the scattering along any given direction.

Now <sup>when  $l=L$</sup>  each of the sums  $\sum_{\xi=-\infty}^{+\infty} \sin^2 \xi / \xi^2$ , etc. appearing in (15) is equal to 2, which is just twice the value of  $\sin^2 \xi / \xi^2$  at  $\xi=0$ , etc. Their product will therefore be 8 times the value at  $\xi=\eta=\zeta=0$ . Now in reflection from a stratified medium, it will be seen by applying Huyghens's Principle, that the variations in density along the normal to the ~~plane of reflection~~ <sup>no plane</sup> alone will affect the intensity of reflection in the Bragg direction. Hence the above result indicates that  $S_{\varphi}$  is eight times the coefficient of reflection along the Bragg direction of reflection from a pair of progressive elastic waves, extending over the whole volume  $L^3$  of the liquid, the two progressive waves travelling in opposite directions along the bisector of the exterior angle between the directions of incidence and of observation, the wave-length of these waves,  $\Lambda_0 = 2L / (\rho_0^2 + \sigma_0^2 + \tau_0^2)^{1/2} = \lambda / (2 \sin \varphi/2)$ , being that appropriate for giving a Bragg reflection of the incident light-waves of wave-length  $\lambda$  along the direction of observation, and the energy of each of the two progressive waves being one-eighth of the energy associated with a normal mode of vibration\*, or one-eighth of the energy of a Planck oscillator of frequency  $N_0$  corresponding to the wave-length  $\Lambda_0$ . It follows that  $S_{\varphi}$  may also be taken to be just the coefficient of reflection along the Bragg direction of reflec-

---

\* since eight such progressive waves constitute one normal mode of vibration.

tion, from these two progressive waves, provided we assign to each of these waves the full energy of a Planck oscillator of the appropriate frequency  $N_0$ .

5. INTENSITY OF SCATTERING IN TERMS OF THE ENERGY AND VELOCITY OF PROPAGATION OF THE REFLECTING ELASTIC WAVES

In the last section we noticed that practically the whole of the contribution to the observed scattering along any given direction comes from the few elastic waves in the close neighbourhood of those in a position to reflect, in the Bragg sense, the incident light waves along the direction of observation, i.e. from those elastic waves whose  $e\sigma\tau$  values lie in the close neighbourhood of  $e_0\sigma_0\tau_0$ , and hence expression (8) for  $S_\varphi$ , namely

$$S_\varphi = n^2 \sigma_\varphi \cdot \frac{\overline{B^2} L^3}{8 D_0^2},$$

will be valid even under conditions when  $\overline{B^2}_{e\sigma\tau}$  may not be independent of  $e\sigma\tau$ , e.g. at low temperatures, provided we use for  $\overline{B^2}$  the value appropriate for  $e_0\sigma_0\tau_0$ . In that case it will be convenient to express  $\overline{B^2}_{e_0\sigma_0\tau_0}$  in terms of the energy and the velocity of propagation of the elastic waves defined by  $e_0\sigma_0\tau_0$ , instead of in terms of  $RT\beta$  as we did at the high temperatures. The expression for  $S_\varphi$  will now be of the form

$$S_\varphi = n^2 \sigma_\varphi \cdot \frac{E(N_0)}{V_0^2 D_0}, \quad \dots (13)$$

where ~~whose~~  $E(N_0)$  is the energy of a Planck oscillator of frequency  $N_0$  and is equal to  $hN_0 / (e^{hN_0/RT} - 1) + \frac{1}{2} hN_0$ , and  $V_0$  is the velocity of propagation of elastic waves of this frequency in the liquid,

$$V_0 / N_0 = \Lambda_0 = 2L / (e_0^2 + \sigma_0^2 + \tau_0^2)^{1/2}.$$

For the long elastic waves  $\Lambda_0 \gg n^{-1/3}$  involved in light-scatter-

ing,  $V_0$  will be practically independent of the wave-length  $\lambda_0$ , and <sup>will be</sup> equal to  $(\beta D_0)^{-1/2}$ , leading to the same expression for  $S_{\text{el}}$  as before.

We have confined ourselves till now to a fluid medium, in which the elastic waves concerned in reflecting the incident <sup>light</sup> waves, and thus producing the observed scattering, are longitudinal waves, the direction of displacement of the atoms under ~~the~~ these waves being along the wave-normal, i.e. along the normal to the <sup>reflecting</sup> plane of reflection. This will be the case in an elastically <sup>(isotropic solid also. If, however, the medium is an elastically</sup> anisotropic solid, the wave-length  $\lambda_0$  and the direction of the wave-normal, say  $\vec{n}_0$  of the elastic waves which by reflection of the incident light waves produce the scattering, will still be determined in the same manner as before, namely by  $\rho_0 \sigma_0 \tau_0$ , but the frequency  $N_0$ , and the velocity of propagation  $V_0$ , of these elastic waves will now be dependent on the direction of ~~propagation~~ <sup>-normal  $\vec{n}_0$</sup>  of the waves in the crystal, and will have three different values, corresponding to the three directions of displacement in the crystal associated with elastic waves of a given wave-length, and given wave-normal direction.   
 $\leftarrow$  Further, the directions of displacement will not in general be parallel and perpendicular respectively to the direction of the wave-normal  $\vec{n}_0$ , as in an elastically isotropic medium, but will be inclined to this direction. Let  $\omega_1, \omega_2, \omega_3$  be the angles which the three directions of displacement make with the direction of the wave-normal of the elastic waves. According to ~~the~~ Huyghens's Principle it is only the components of the displacements along the normal to the <sup>reflecting</sup> plane of reflection that will <sup>determine</sup> ~~affect~~ the intensity of reflection. Hence the ~~expression for the total scattering of all the Brillouin components~~ <sup>coefficient  $S_{\text{el}}$</sup>  will now be given by

$$S_{\text{el}} = \frac{n^2 \sigma_{\text{el}}}{D_0} \sum_{i=1}^3 \frac{E(N_i) \cos^2 \omega_i}{V_i^2}, \quad \dots (18)$$

determine

where  $N_i = V_i / \Lambda_0$ , and  $V_i$ ,  $i=1, 2, 3$ , are the frequencies and the velocities of propagation of the three polarized elastic waves associated with the wave-length  $\Lambda_0$ , and the wave normal  $\vec{n}_0$ , both defined by  $\epsilon_0 \sigma_0 \tau_0$ .

Expression (18) can be readily recognised as Waller's (1925) expression for the intensity of scattering of X-rays of wave-length  $\lambda$  <sup>for the case</sup> when  $\Lambda_0 = \lambda / (2 \sin \frac{\varphi}{2}) \gg n^{-1/2}$ .  $V_i$  can be expressed as usual in terms of the elastic constants of the crystal, which for such long wave-lengths  $\Lambda_0$  as are involved here, will be practically the same as the static elastic constants of the crystal.

## 6. THE ATOMIC CROSS-SECTION FOR SCATTERING

Coming back to the liquid medium, we proceed to evaluate the cross-section of the atom in the liquid for scattering, namely  $\sigma_{\varphi}$ . If the incident light is linearly polarized, and the direction of observation makes an angle  $\theta$  with the electric vector of the incident light ( and  $\varphi$  with the direction of propagation of the incident light, as before )  $\sigma_{\varphi}$ , as we have defined it, will evidently be given by

$$\sigma_{\varphi} = (2\pi/\lambda)^4 M^2 \sin^2 \theta, \dots (19)$$

where  $M$  is the dipole moment induced in the atom per unit "field in the medium", as usually defined, of the electric vector of the incident light-wave, and is given by

$$M = \chi/n = \frac{\epsilon - 1}{4\pi n}, \dots (20)$$

where  $\chi$  is the optical susceptibility and  $\epsilon$  is the

square of the refractive index, of the liquid.

Hence

$$\sigma_{\varphi} = \frac{\pi^2 (\epsilon - 1)^2}{n^2 \lambda^4} \sin^2 \theta. \quad \dots(21)$$

We should emphasise here that the field actually acting on the atom, and producing the dipole moment  $M$ , is not merely the field in the medium, say  $E$ , but also includes the polarization field  $P$  due to the dipole moments similarly induced in all the surrounding atoms. Hence  $\chi$  in expression (20) will not be just proportional to  $n$ , (except in a gas where  $\epsilon \sim 1$ ), but will be greater. If the polarization field  $P$  has the Lorentz value, namely

$4\pi\chi E/3$ , the total field which acts on an atom and induces the dipole moment  $M$ , will be  $1 + 4\pi\chi/3$  or  $(\epsilon + 2)/3$  times  $E$ , where  $\epsilon$  is the mean dielectric constant of the medium. Hence neither  $M$ , nor  $\sigma_{\varphi}$  as we have defined it, which is proportional to  $M^2$ , will be atomic constants, but will depend also on the density of packing of the atoms in the medium. If the dielectric constant conforms to the Lorentz formula,  $\sigma_{\varphi}$  will be proportional to  $(\frac{\epsilon + 2}{3})^2$ , and will be correspondingly much higher in the liquid than in the gaseous state.

Expressions (19) and (20) are intended to refer to the case when the incident light is linearly polarized, and the direction of observation makes an angle  $\theta$  with the direction of the electric vector of the incident light wave. If the incident light is unpolarized,  $\sin^2 \theta$  in the above expressions will have to be replaced by  $(1 + \cos^2 \theta)/2$ , and for other polarizations of the incident light by a suitable factor  $f(P)$  which can be readily calculated.

Substituting the value of  $\sigma_{\varphi}$  deduced here in expression (10) for  $S_{\varphi}$ , we obtain

$$S_{\varphi} = \frac{\pi^2 k T \beta}{\lambda^4} (\epsilon - 1)^2 f(P). \quad \dots(22)$$

7. COMPARISON WITH EINSTEIN'S VALUE

We expressed  $S_{\epsilon}$  in the first place in terms of the atomic scattering coefficient  $\sigma_{\epsilon}$ , and thence deduced expression (22) for  $S_{\epsilon}$ , whereas in Einstein's derivation, as we mentioned in an earlier section,  $S_{\epsilon}$  is connected directly with the local fluctuations in the refractivity of the liquid accompanying the fluctuations in density. The two are related in the following manner.

$n^2 \sigma_{\epsilon}$  in our notation gives the scattering per unit volume from an element of volume when all the atoms in the element scatter in the same phase. This would correspond in Einstein's derivation to  $\frac{\pi^2}{\lambda^4} \cdot (D \frac{\partial \epsilon}{\partial D})^2 \frac{1}{\lambda^2}$ . In evaluating  $D \frac{\partial \epsilon}{\partial D}$  Einstein adopts the Lorentz relation

$$\frac{\epsilon - 1}{\epsilon + 2} \cdot \frac{1}{D} = \text{constant}, \quad \dots(23)$$

which gives on differentiating with reference to D

$$D \frac{\partial \epsilon}{\partial D} = (\epsilon - 1) \frac{\epsilon + 2}{3}, \quad \dots(24)$$

which leads to Einstein's expression for  $S_{\epsilon}$ , namely

$$S_{\epsilon} = \frac{\pi^2 k T \beta}{\lambda^4} (\epsilon - 1)^2 \left(\frac{\epsilon + 2}{3}\right)^2 f(P), \dots(25)$$

This differs from (22) in that it includes the factor  $\left(\frac{\epsilon + 2}{3}\right)^2$  which is not present in (22).

This discrepancy is due to the following circumstances.  $\partial \epsilon / \partial D$  occurring in Einstein's expression for  $S_{\epsilon}$  denotes the fluctuation in  $\epsilon$  in any small element of volume of the liquid accompanying the fluctuation in its density  $\frac{D}{\lambda}$  per unit value of the latter. Adopting a suitable formula connecting  $\epsilon$  and  $D$  for the liquid, for example the Lorentz formula (23), as we have done, we would not be just justified in obtaining the required value of  $\partial \epsilon / \partial D$  by differentiating  $\epsilon$  in the expression with reference to  $D$ . Such a differentiation would give the change in  $\epsilon$  consequent on a change of D, per unit value of the latter, when the density D of the liquid is varied uniformly ~~throu~~

throughout the liquid, whereas the variations of density we are concerned with here are local fluctuations, the fluctuations in different regions being uncorrelated with one another. In order to obtain the value of  $\partial\epsilon/\partial\mathcal{D}$  appropriate for such local variations in density, we need to know how the polarization field at any point, which is equal to  $4\pi\chi/3 = (\epsilon-1)/3$  times the field in the medium, and which together with the latter gives a total field  $(\epsilon+2)/3$  times the field in the medium, varies with the fluctuations of density at the point. Now the polarization field at any point will be determined not only by the density of distribution of the atomic dipoles in its close neighbourhood but in the whole of the surrounding regions. Hence though  $\epsilon$  appearing in the numerator of (23) will vary in accordance with the fluctuations in density at the point,  $\epsilon$  occurring in the denominator of (23), which comes through the factor  $(\epsilon+2)/3$ , which gives the ratio of the actual field to the field in the medium, both at the point, will not vary in the same manner. From some arguments put forward by Ramaathan (1927) it appears that to a first approximation the polarization field at any point in the medium should be independent of the fluctuation in density at the point. In other words the value of  $\partial\epsilon/\partial\mathcal{D}$  appropriate to our problem would approximate more closely to the expression

$$\mathcal{D} \frac{\partial\epsilon}{\partial\mathcal{D}} = \epsilon - 1, \quad \dots (26)$$

rather than to

$$\mathcal{D} \frac{\partial\epsilon}{\partial\mathcal{D}} = (\epsilon - 1) \frac{\epsilon + 2}{3}$$

obtained by direct differentiation of Lorentz's expression, and given in (24).

The fundamental assumption underlying our derivation of expression (3) for  $S_{\mathcal{D}}$ , namely that the contribution to  $S_{\mathcal{D}}$  from a small element of volume  $v$  in the liquid containing on an average  $N$  atoms, and so small that the

scattered radiations from different parts of the volume may be regarded as being in practically the same phase, is given by ~~see~~ (6)  $S_{\varphi} \nu = \overline{\Delta N^2} \sigma_{\varphi}$ , is indeed equivalent to assuming that the Lorentz polarization field at any point in the liquid is practically independent of the local fluctuation of density at the point. Hence the absence of the factor  $\left(\frac{\epsilon+2}{3}\right)^2$  in our expression for  $S_{\varphi}$ , as compared with Einstein's.

The available experimental data for light-scattering in a large number of liquids, as was shown by one of us several years ago (Krishnan, 1925), point definitely to the need for deleting the factor  $\left(\frac{\epsilon+2}{3}\right)^2$  from Einstein's expression (20) for the scattering coefficient in a liquid.

#### 8. BRILLOUIN SPLITTING IN LIGHT-SCATTERING

We have neglected till now the small changes in frequency accompanying scattering, i.e. the enhancement and degradation in frequency accompanying the reflections respectively from the two sets of progressive waves travelling in opposite directions, to which attention was first drawn by Brillouin (1914, 1925). Consider an

elastic wave defined by  $e^{\sigma\tau}$  and having a frequency  $N_{\sigma\tau}$ .

(and extending over the whole volume  $L^3$  of the liquid.)

The contribution from this wave to the scattering coefficient  $S_{\varphi}$  along the direction  $\varphi$  selected for observation of scattering, which we shall denote by  $[S_{\varphi}]_{\sigma\tau}$  will now consist of two terms, as Brillouin (1933) has shown, namely

$$[S_{\varphi}]_{\sigma\tau} = [S_{\varphi}]_{\sigma\tau}^{+} + [S_{\varphi}]_{\sigma\tau}^{-}, \quad \dots(27)$$

where the two terms with superscripts + and - correspond respectively to scattering of enhanced and degraded

delete 11

frequencies, namely  $\nu' = \nu \pm N_{\rho\sigma\tau}$ . Let us denote by  $k_x, k_y$  and  $k_z$  the components of the wave-vector  $\vec{k}$  of the incident light-wave, and by  $k'_x, k'_y$  and  $k'_z$  the components of the wave-vector  $\vec{k}'$  of the scattered radiation;  $|k| = 1/\lambda = \nu/v$ , and  $|k'| = 1/\lambda' = \nu'/v'$ , where  $v$  and  $v'$  are the velocities in the liquid of the incident and the scattered radiations respectively;  $v'$  besides being slightly different from  $v$ , will also be slightly different for the two values of  $k'$ . Either of the two terms in (27) will now be given by

$$[S_{\rho\sigma\tau}]_{\rho\sigma\tau}^{\pm} = n^2 \sigma_{\rho} \frac{L^3}{128 D_0^2} \overline{B_{\rho\sigma\tau}^2} \frac{\sin^2 \xi}{\xi^2} \cdot \frac{\sin^2 \eta}{\eta^2} \cdot \frac{\sin^2 \zeta}{\zeta^2}, \quad \dots (28)$$

in which  $\xi, \eta, \zeta$  are now given by

$$\left. \begin{aligned} \xi &= \frac{\pi}{2} \rho - \pi L |k_x - k'_x| \\ \eta &= \frac{\pi}{2} \sigma - \pi L |k_y - k'_y| \\ \zeta &= \frac{\pi}{2} \tau - \pi L |k_z - k'_z| \end{aligned} \right\} \dots (29)$$

$[S_{\rho\sigma\tau}]_{\rho\sigma\tau}^{+}$  and  $[S_{\rho\sigma\tau}]_{\rho\sigma\tau}^{-}$  corresponding respectively to the two values of  $k'$ .

It will be seen from (29) that the condition  $\xi = \eta = \zeta = 0$ , which defines the directions of maximum intensity of reflection for the two frequencies, is not now the same as the Bragg condition, namely  $\rho = \rho_0, \sigma = \sigma_0$  and  $\tau = \tau_0$ , but corresponds to slightly different values of  $\rho, \sigma, \tau$ , which we shall denote by  $\rho', \sigma', \tau'$  respectively.  $\rho', \sigma', \tau'$  will also be slightly different for the two Brillouin components.

In the special case when  $k'$  is put equal to  $k$ , i.e., when we neglect the change in frequency accompanying

reflection,  $e'_0 \sigma'_0 \tau'_0$  reduce to  $e_0 \sigma_0 \tau_0$  as they should, and  $[S_\varphi]_{e\sigma\tau}^+$  will then be equal to  $[S_\varphi]_{e\sigma\tau}^-$ , and  $[S_\varphi]_{e\sigma\tau}$  will be twice the value given by the right hand side of (28).

Considering the general case, and confining ourselves to a given direction of incidence, and a given direction of observation, it will be readily seen from (19) that as we pass from one elastic wave to another, the corresponding change in  $\xi$  (and similarly in  $\eta$  or  $\zeta$ ) will be given by

$$\Delta \xi / \Delta e = \pi/2 + \pi L \cdot \Delta k'_x / \Delta e, \quad \dots(30)$$

which in view of the relation  $K_x = e/(2L)$ , where  $K_x$  is the  $x$ -component of the elastic wave-vector  $\vec{K}$ , and

*delete 1)*  $|\vec{K}| = (e^2 + \sigma^2 + \tau^2)^{1/2} / (2L)$ , becomes

$$\frac{\Delta \xi}{\Delta e} = \frac{\pi}{2} \left[ 1 + \frac{\Delta k'_x}{\Delta K_x} \right] = \frac{\pi}{2} \left[ 1 + \frac{k'_x}{k'} \cdot \frac{K_x}{K} \cdot \frac{\Delta k'}{\Delta K} \right] \dots(31)$$

Now

$$\frac{\Delta(v'k')}{\Delta e} = \pm \frac{\Delta N}{\Delta e} = \pm \frac{\Delta(VK)}{\Delta e} \dots(32)$$

Hence

$$\frac{\Delta k'}{\Delta K} = \pm \frac{\Delta N / \Delta K}{\Delta v' / \Delta k'} = \pm \frac{G}{g'}, \quad \dots(33)$$

where  $g'$  and  $G$  are the group velocities of the scattered light-waves, and of the elastic waves in the neighbourhood of the wave-number  $K$ , respectively.

$$\therefore \frac{\Delta \xi}{\Delta e} = \frac{\pi}{2} \left[ 1 \pm \frac{k'_x}{k'} \cdot \frac{K_x}{K} \cdot \frac{G}{g'} \right] \dots(34)$$

In the second term inside the square brackets,  $k'_x/k'$  and  $K_x/K$  are direction-cosines, and hence will be less than unity,

not exceed  
~~than~~ unity, and  $G/g'$  will in general be of the same order of magnitude as  $v/v'$ , the ratio of the velocity of sound to that of light, and hence the second term will be negligible in comparison with the first term, and  $\Delta\xi/\Delta\rho$ , and similarly  $\Delta\eta/\Delta\sigma$  and  $\Delta\zeta/\Delta\tau$ , will be almost exactly equal to  $\pi/2$ . In other words as we pass from one stationary elastic wave to the adjacent one, and the  $\rho$  or  $\sigma$  or  $\tau$  values change by steps of unity, the corresponding changes in  $\xi$   $\eta$   $\zeta$  will be in steps, <sup>of</sup> practically of  $\pi/2$  as before.

Since  $\Delta\xi/\Delta\rho$ , etc., are practically  $\pi/2$ ,  $[S_\varphi]_{\rho\sigma\tau}^+$  or  $[S_\varphi]_{\rho\sigma\tau}^-$  will be significant for only a few values of  $\rho\sigma\tau$  in the close neighbourhood of the corresponding  $\rho'_0\sigma'_0\tau'_0$ , or practically of  $\rho_0\sigma_0\tau_0$ , since  $\rho'_0 - \rho_0$ , etc. can be readily shown to be less than  $4Lk \cdot v/v'$ , and hence less than 1 when  $L$  is of the order of unity. In other words the frequencies  $N_{\rho\sigma\tau}$  and the mean square of amplitude  $\overline{B_{\rho\sigma\tau}^2}$ , for the elastic waves concerned effectively in scattering either of the Brillouin components, will be practically those corresponding to  $\rho_0\sigma_0\tau_0$ . In other words for any given direction of incidence, and of observation at an angle  $\varphi$ , the changes in frequency will be practically  $\pm N_0$ , and the intensity of either component will be given by

$$S_\varphi^+ = S_\varphi^- = n^2 \sigma_\varphi \frac{L^3 \overline{B^2}}{128 D_0^2} \sum_{\rho} \sum_{\sigma} \sum_{\tau} \frac{\sin^2 \xi}{\xi^2} \cdot \frac{\sin^2 \eta}{\eta^2} \cdot \frac{\sin^2 \zeta}{\zeta^2} \dots (35)$$

$$= n^2 \sigma_\varphi \cdot \frac{L^3 \overline{B^2}}{16 D_0^2}$$

$$= n^2 \sigma_\varphi \cdot \frac{E(N_0)}{2V D_0} \dots (36)$$

Thus the coefficient of scattering of either of the Brillouin components separately, of enhanced or degraded frequency,  $\nu' = \nu \pm N_0$  per unit volume, per unit solid angle, will as before be equal to eight times the maximum

intensity of reflection from a progressive wave of the appropriate frequency  $N_0$  travelling along the appropriate direction, and extending over the whole volume  $L^3$  of the liquid, and having <sup>one eighth</sup> the energy of <sup>a</sup> Planck oscillator of frequency  $N_0$ .

This result is the same as that obtained by Brillouin (1933) who assumed the two progressive waves that give rise respectively to the two spectral components in scattering as each having the full energy of a Planck oscillator, and further arbitrarily took the scattering coefficient as equal to the reflection coefficient at the maximum corresponding to  $\xi = \eta = \zeta = 0$ . \*

In a crystal too, the wavenumbers  $K'_0$  of the elastic waves which effectively contribute to <sup>the</sup> scattering along any given direction, will be defined as before by  $\rho'_0 \sigma'_0 \tau'_0$ , and practically by  $\rho_0 \sigma_0 \tau_0$ , but in general there will be three different values of  $N_0$  associated with the three velocities of propagation of a wave whose wave-length and wave-normal are defined by  $\rho_0 \sigma_0 \tau_0$ , and hence there will be three pairs of Brillouin components whose frequencies are

$$\nu'_i = \nu \pm N_i ; \quad i = 1, 2, 3.$$

On this basis the intensities of <sup>the</sup> Brillouin components corresponding to the same frequency shift on either side, will be ~~of~~ the same ~~intensity~~, whereas the intensities of the three components on any one side, corresponding to the three values of  $N_i$ , will be widely different, the intensities of the three being given respectively by (See (18))

$$S_{\varphi i}^{\pm} = \frac{n^2 \tau_{\varphi}}{2D_0^2} \cdot \frac{E(N_i) \cos^2 \omega_i}{V_i^2} ; \quad i = 1, 2, 3, \dots (37)$$

and  $\omega_i$  and  $N_i$ , and hence  $V_i$  also, being widely different for the three components.

9. RELATIVE INTENSITIES OF THE STOKES AND THE  
ANTI-STOKES BRILLOUIN COMPONENTS

From the point of view adopted above, which regards the Brillouin components on either side as Doppler shifts accompanying reflections from the appropriate progressive waves moving in opposite directions, the enhanced and degraded frequencies corresponding to the same frequency - shift  $N_i$  should be of the same intensity. Though this will be true at all ordinary temperatures  $T \gg \hbar N_i / k$ , where  $k$  is the Boltzmann constant, this will not be the case at low temperatures. The intensities will then have to be calculated on the quantum theory, as in the case of the intensities of the Raman spectra. Indeed the Brillouin components are the Stokes and the anti-Stokes Raman lines respectively, due to acoustic frequencies, whereas the usual Raman spectra are due to the vibrational and rotational frequencies characteristic of the medium, i.e., due to the optical branches of the elastic waves, which also will be present in general, and which for simplicity, we have neglected till now, in the crystal explicitly, and in the liquid implicitly by assuming the liquid to be monatomic.

There is one striking difference between Raman spectra due to the acoustic and the optical branches of the elastic waves. Whereas for the optical branch of the elastic waves the frequency  $N_i$  is practically independent of the wave-length  $\Lambda = \lambda / (2 \sin \frac{\theta}{2})$ , and hence the corresponding Raman shifts  $\Delta \nu_i = \pm N_i$  are independent of  $\Lambda$ , and therefore of the direction of scattering, for the acoustic branch  $N_i$  is nearly proportional to  $1/\Lambda$ , and hence the frequency shifts of the Brillouin components are proportional to  $1/\Lambda$ , and hence vary as  $\sin \frac{\theta}{2}$ .

The effect of including the optical branch will be two-fold. Besides the appearance of the usual Raman spectra

the polarization characteristics of ordinary scattering also will be greatly altered, since  $\sigma_{ce}$  for the scattering <sup>atoms</sup> ~~units~~ will be a function of the direction of the electric vector of the incident light wave in relation to the dispositions of the different atoms. In other words the optical branch of the elastic spectrum will not only produce Raman spectra corresponding to much larger frequency shifts than are involved in Brillouin splitting, but will affect the polarization of ordinary scattering — in a liquid the effect will be the same as that attributed in the usual treatment to the optical anisotropy of the molecules in the medium.

#### S U M M A R Y

The intensity of light scattered by a homogeneous monatomic liquid can be calculated readily, following Einstein, by attributing the scattering to the local fluctuations in density, and regarding the latter as due to the superposition of the standing or progressive elastic waves of different wave-lengths. The expression for the scattering coefficient along any given direction involves a triple infinite series, each of which is of the form  $\sum_{n=-\infty}^{+\infty} \alpha \frac{\sin^2(n\alpha + \theta)}{(n\alpha + \theta)^2}$ , where  $n$  is an integer, and  $\alpha$  and  $\theta$  are positive constants. By splitting the medium into small volume elements,  $\alpha$  may be made sufficiently small and the summation may be replaced by integration, and this is usually done. But this is not necessary, since the series can be summed even when  $\alpha$  is not so small, provided  $0 < \alpha \leq \pi$ , which is actually the case, since, even when the volume element chosen is the whole of the medium  $\alpha$  is just  $\pi/2$ .

Taking the medium as a whole, one finds that the elastic waves regarded as forming a reflection grating, have a high resolving power, actually half that necessary

to resolve in the Rayleigh sense the reflections from adjacent elastic waves; and hence it becomes convenient to regard scattering in terms of reflection from the appropriate elastic waves.

On this basis, the coefficient of scattering along any given direction comes out to be eight times the coefficient of reflection at the maximum from just two progressive waves in the medium which satisfy the Bragg law, each of the two waves having one-eighth the energy of a Planck oscillator of the same frequency as the waves.

These two progressive waves give rise respectively to the two Brillouin components in scattering, and the intensity of either of them deduced in the above manner agrees with that <sup>given</sup> ~~calculated~~ by Brillouin, who assumed each of the progressive waves to have the full energy of a Planck oscillator, and who further arbitrarily took the scattering coefficient as equal to the reflection coefficient at its maximum.

The calculation of intensity is extended to a crystalline medium, and the expression for the total intensity of all the Brillouin components together, is shown to be identical with Waller's expression for X-ray scattering.

For more precise calculation of intensity the Brillouin components have to be regarded as the Raman spectra due to acoustic frequencies, as distinguished from the usual Raman spectra, which also will be present when the elastic spectrum consists of optical branches also.

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