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# Selected papers of Raman: An introduction

T. V. RAMAKRISHNAN
Department of Physics, Indian Institute of Science, Bangalore 560 012.

A small selection of papers by Professor C. V. Raman follows this introduction. Some have been selected for their immediate as well as lasting impact, others for their prescience. They illustrate his characteristically direct and incisive approach, his interest in natural phenomena, his concern for basic questions and implications, and his eye (and ear) for beauty.

Raman's writing style is direct, clear and has a characteristic literary flavour. For these reasons annotating his papers is often an undesirable intrusion. However, many of them are very brief, and were written in a certain intellectual milieu for a particular audience. Several papers hint at areas of physics which have seen growth much later. In this introduction, I try therefore to expand on the matter condensed in a few lines, to set out the background, or to point out how the work foreshadows present concerns in physics.

The very first paper [P1] is an extremely brief and incomplete summary of one of Raman's two major contributions to musical acoustics, namely his work on the Mṛdangam (and the Tabla). These Indian drums are true musical instruments, the musical quality being due to the harmonic overtones and their relative intensities, as well as to the clarity, duration and volume of sound. Now in a simple circular drum, the overtones are not integral multiples of a fundamental or lowest frequency mode of vibration. It is therefore not musical. Raman guessed that the Indian drum is designed (with its nonuniform black centre and an auxiliary circular leather strip) to produce harmonic overtones, to damp out high-frequency harmonics and to produce clear long-lasting notes. He showed this by exciting various harmonics, and determined their vibration patterns by noting the places where sand, spread on the drum, collects. (It collects at places which do not vibrate, called nodes; their number and shape are diagnostic of the frequency and nature of the mode of vibration). This note does not detail or even describe these results. They were written up much later, in 1934 [The Indian musical drums, *Proc. Indian Acad. Sci.*, 1934, **1**, 179–188], though all the major results had been obtained before 1921. The reason for this delay was Raman's increasing preoccupation with light and its scattering, the beginnings of which can be seen in the next paper. Anyone interested in the area of musical acoustics should read his 1934 paper. It describes Raman's discoveries in detail, with pictures of the drum in various 'pure' modes of vibration.

The second paper, on 'The colour of the sea' [P2] is a Raman classic. A great scientist, Lord Rayleigh (whom Raman particularly admired) said, perhaps without thinking much about it, that the blue of the sea is just the reflected blue of the sky. Raman, on his first voyage abroad, disposed of this suggestion quickly with a few deckside observations using a Nicol prism analyzer. The idea is the following: the light reflected from water is partially polarized, the polarization being nearly complete for a certain angle of incidence. The Nicol prism, if oriented in a particular direction, will completely block out polarized light. By this simple arrangement, reflected light can be eliminated. Raman found, under these conditions, that the blue of the sky instead of disappearing was 'wonderfully improved'! His belief that the blue of the ocean had the same physical cause as the blue of the sky (explained by Lord Rayleigh!) was reinforced by two observations. The first is that when the reflected light is cut out, the colour of the sea depends on the angle between the incident sunlight and the sea-scattered light reaching the eye. Further, the track of sunlight in water could be seen under certain conditions out to considerable depth. Now, Lord Rayleigh had argued that the blue of the sky is due to scattering of sunlight in the following way. Purely statistically, at a given instant some regions of air are denser, some others rarer. Since the refractive index of air depends on density, these spontaneous density fluctuations mean that the refractive index fluctuates randomly. This scatters sunlight; the extent or intensity of scattering depends on angle as well as on light wavelength, increasing inversely as the fourth power of the latter. Thus the blue part of sunlight is scattered much more than the longer wavelength parts. There is every reason to expect that similar fluctuations in density should occur in water as well and so the same scattering process should be operative. Raman pursued this idea vigorously, and produced a fairly detailed analysis of the colour of the sea rather soon (*Proc. R. Soc.*, 1922, **A101**, 64–80, and a monograph entitled 'Molecular diffraction of light' published by the Calcutta University in 1922). The point is that this scattering is due not to extraneous causes such as dust or other substances, but is due to density fluctuations.

Raman's sustained efforts at observing this Rayleigh scattering in dust-free fluids in the laboratory originate with this experience. These bore fruit in several spectacular ways. Some of these are subjects of the next few reprints.

Raman was probably the first person to appreciate the implications of light scattering from glassy or amorphous solids, and viscous liquids. Glasses are generally made by rapidly cooling melts or liquids, the cooling rate being large enough to prevent crystallization or formation of the crystal. If the arrangement of atoms in the glass is just the same as in the melt, but is frozen-in, then light would be scattered from the frozen-in density fluctuations appropriate to that for the melt *i.e.* the liquid. Another possibility is that atoms in a glass are positionally well ordered, almost as in the corresponding crystal, but that the constituent anisotropic molecules are not orientationally ordered, *i.e.* there are considerable orientational fluctuations (frozen-in again). A third possibility is that especially in chemically complex glasses (*e.g.* optical or window glass) there are sizeable compositional fluctuations. In general, things tend to separate out at low temperatures (entropy is less at lower temperatures!) so that these compositional fluctuations tend to increase on cooling. In a particular glass, all these effects could be present to different degrees at different length (and time) scales.

In the very first brief paper on the subject [P3] Raman argued, from the rather large Rayleigh-like scattering of light in glasses, that there are sizeable frozen-in density fluctuations in them, comparable to those in liquids. Thus a quasicrystalline model for the glassy state, with sizeable crystallites, is ruled out. To confirm for himself the hypothesis that the scattering of light from glasses is due to intrinsic fluctuations and not accidental 'inclusions' and inhomogeneities, Raman took a dozen optical glasses with graded refractive indices and studied the intensity and the depolarization of light scattered from these [P11]. He found that the former increases and the latter decreases as the refractive index increases. This systematic dependence suggests that the scattering process at work is intrinsic, related to their 'optical density' and is not extrinsic. These contributions mark the beginning of a powerful way of probing a class of systems and phenomena not fully understood yet.

Glasses are strange systems which have fallen out of thermal equilibrium in regard to some degrees of freedom (or configurations) and are in equilibrium as far as other degrees of freedom (*e.g.* local atomic vibrations) are concerned. That is, the distribution of energy in some arrangements of atoms or molecules cannot be described by a temperature, while for some other arrangements it can be! The dynamics of this process, *i.e.* how it occurs, is an area of great current interest. Light-scattering techniques are much more sophisticated now and time-dependent processes are routinely studied. Fascinating very slow relaxation processes have been uncovered. However, one still does not have a deep understanding of these phenomena which are common to all glasses.

Another contribution in the same general area is light scattering from viscous liquids [P9]. Raman was fascinated by the following idea, due primarily to Brillouin. The thermal (temperature-induced) agitation of atoms in a liquid (more clearly in a solid) can be thought of a sum (or superposition) of sound waves of different wavelengths, amplitudes and directions of propagation. Scattering of light from one such wave will lead to a light wave with a slightly different (shifted) direction of propagation and nearly the same frequency just as happens when light is incident on a diffraction grating. This Brillouin line was looked for by Raman and coworkers. Now if the sound wave damps out quickly, such a distinct line won't be seen. The damping is due to viscous friction in a liquid, and will thus decrease as temperature increases (and fluid viscosity decreases). Thus on increasing the temperature of a liquid, the Brillouin line should appear; this was exactly what Raman and Raghavendra Rao found. This also is an active current area of research; one would like to probe and understand the way the Brillouin line disappears as one approaches the glass transition marked by a catastrophic viscosity increase.

The paper on optical behaviour of protein solutions [P5] is again well ahead of its time. Whereas large protein molecules in solution were thought of as particles of dust leading to haze or light scattering, Raman considered them to be a gas of particles with a temperature, compressibility, etc. It turns out that this gas or fluid changes its nature (*e.g.* from gas to liquid) as its density, or the medium in which the molecules are suspended, changes. At such a transition point, fluctuations in density are very prominent, and we expect the same excess scattering of light as occurs for example at the liquefaction point of carbon dioxide. The study of phase changes in polymers, colloids and gels has emerged as a major field and light scattering is an important tool for monitoring molecular movements, arrangements and cooperative changes. This was probably the first paper in which one such object (protein solution) was thought of as a thermodynamic system.

Raman's interest in understanding the arrangement of molecules in dense systems such as liquids led to the pioneering use of X-ray scattering (diffraction) for probing their structure. Raman and Ramanathan had realized in 1923 that since X-rays have wavelengths of molecular dimensions, small-angle scattering is related to the statistical density fluctuations described earlier but large-angle scattering is a direct probe of local molecular arrangement. This large-angle scattering shows up as haloes [P4]. Raman and Sogani compared the haloes for two organic compounds, hexane and cyclohexane, and argued that the differences are due to specific differences in molecular shape. This is the first work on X-ray diffraction as a structural tool in liquids. The work would have been complete if the authors had realized that the Fourier transform $g(r)$ of the observed intensity $S(q)$ (as a function of wavevector change $q$) gives the two-particle correlation function or the probability of finding two molecules a distance $r$ apart, a quantity of direct physical appeal. This was done by Zernicke and Prins in 1927.

The papers P6, P7 and P12 describe the most celebrated contribution of Raman to science, namely the Raman Effect. Raman, his students, and coworkers had been investigating the scattering of light from liquids since 1921 or so. Most observations, some of which have been described above, could be explained by assuming light to be a classical wave scattered by spontaneous fluctuations in the medium. However one persistent phenomenon did not fit into this pattern; it was dubbed 'weak fluorescence' to begin with. Clues about its nature and evidence for its general occurrence, were obtained over the years. One such is the following. By using a pair of complementary filters, *i.e.*, filters such that one filter blocks the colours that the other transmits, Raman and Krishnan found the following: When both the filters are placed in the path of the beam incident on the pure fluid no scattered light is seen. However, if one filter is placed in the path of the incident beam and the other in the path of the scattered beam, some weak fluorescence could be seen. This clearly means that the colour (or wavelength) of the light transmitted by the first filter changes on scattering by the fluid so that it is no longer exactly complementary to the other filter and is therefore not fully blocked by it. Raman was reinforced in this belief by the then recently discovered Compton Effect in which the wavelength of X-rays hitting a 'free' electron is changed. He reasoned that there could be an optical analogue of the Compton Effect, namely that a light wave changes its wavelength (or frequency or energy) on being scattered by an atom or molecule. To check this, a source emitting light of a single colour or wavelength is needed. An intense mercury vapour lamp with proper filters is such a monochromatic source. When the scattered light was viewed by a spectroscope (in which light of different wavelengths shows up as differently placed lines) Raman found, in addition to the line corresponding to the original wavelength, another line with a shifted (increased) wavelength. In some substances, more than one line was seen; there were also lines with reduced wavelength. The spectra were recorded, and the papers reprinted here show some of them.

Why is this 'new radiation' important? With his uniquely well-prepared mind, Raman understood it all. Firstly, it is striking evidence for the quantum nature of light. A light quantum hits a molecule, and excites one of its internal modes. The remaining electromagnetic energy comes out as a light quantum or photon of reduced energy or increased wavelength. If the light quantum absorbs a quantum of internal molecular excitation, the outcoming photon has higher energy or shorter wavelength. Thus Einstein's ideas (put forward to explain the photoelectric effect) that light energy is carried in quanta, and that this energy is equal to $(hc/\lambda)$ where $h$ is a constant called Planck's constant, $c$ the velocity of light and $\lambda$ the wavelength, are strongly supported. At a more detailed level, Raman realized that the polarizability of the molecule was involved in the mechanism. (This is the same property which causes the refractive index of the liquid to differ from unity and to depend on light wavelength). Indeed, he realized that a detailed theory of molecular and atomic polarizability, due to Kramers and Heisenberg, suggested the possibility of such an effect.

The other major implication, clear to Raman, was that here was a tool of great convenience, precision and power for studying internal excitations of molecules, *e.g.* their vibrations, rotations, and electronic-excited states. Instead of studying the former by direct absorption in the infrared and far-infrared, one could conveniently investigate them in the optical region, as energy differences. Now molecular structure and binding are the backbone of chemistry, so he foresaw that this new branch of spectroscopy would be important for chemistry. With the advent of lasers (monochromatic light sources of great intensity and coherence) Raman spectroscopy became a standard physico-chemical technique.

The availability of lasers has brought about an explosive increase in the number of light-scattering experiments, as several articles in this volume describe. In his Nobel lecture, Raman said: "The universality of the phenomenon, the convenience of the experimental technique and the simplicity of the spectra obtained enable the effect to be used as an experimental aid to the solution of a wide range of problems in physics and chemistry. Indeed, it may be said that it is this fact which constitutes the principal significance of the effect. The frequency differences from the spectra, the width and character of the lines appearing in them, and the intensity and state of polarization of the scattered radiations enable us to obtain an insight into the ultimate structure of the scattering substance. As experimental research has shown, these features in the spectra are very definitely influenced by physical conditions, such as temperature and state of aggregation, by physico-chemical conditions, such as mixture, solution, molecular association and polymerization, and most essentially by chemical constitution. It follows that the new field of spectroscopy has practically unrestricted scope in the study of problems relating to the structure of matter. We may also hope that it will lead us to a fuller understanding of the nature of light, and of the interactions between matter and light". The prophecy has been fulfilled, I think even beyond the dreams of its visionary discoverer.

The paper in this collection on anomalous diamagnetism [P8] is an example of the quickness and richness of Raman's scientific imagination. Diamagnetism is due to electrons circulating in closed (atomic) orbits. Ehrenfest had suggested that the large diamagnetism of bismuth implied that in crystalline Bi electron orbits are specially large. Raman argues in this paper that many facts *e.g.* large change of electrical resistance in magnetic field, anomalous Hall Effect, large change of size in a magnetic field, change of these properties with temperature, are all connected with this. Now with hindsight (as well as the idea of holes and an extremely detailed knowledge of electronic states in Bi) it appears that many of the suggestions are not correct, but let us just look at the quick reach!

The paper with Nagendra Nath [P13] is one of a series of five, in which a phenomenon first observed by several others, was analyzed in a characteristically direct fashion. A high-frequency sound wave propagates in a liquid, and on being reflected by a plane boundary wall, forms stationary waves, or fixed sinusoidal patterns of lower and higher liquid density (and hence refractive index). Now suppose a plane light beam is incident on this. It will be diffracted. The pattern of intensity of diffracted light is observed to be very complex, with a large number of maxima and minima, and considerable wandering of their relative intensities as the angle of light incidence or sound wavelength are varied. Raman and Nath assumed to begin with that the periodic change in refractive index affects only the phase of the plane wavefront sinusoidally. Thus the outcoming wavefront is no longer planar, but corrugated. They calculated diffracted beam intensities, and showed that the complex observations are explained simply. In further work, the assumption that only the phase changes (valid if the light wavelength is much shorter than the sound wavelength) was given up, and amplitude modulation also considered. This whole work is remarkable for its perfection (a well-defined, interesting and rich phenomenon was completely explained theoretically), for its prototypical nature, and because it is a primarily theoretical contribution from a great experimenter. (Contrary to popular belief Raman not only had a sure and creative grasp of theoretical concepts and principles, he was also conversant with theoretical methods, and of course in detail with the great masters *e.g.* Rayleigh, Helmholtz, as well as with the theoretical papers of relevance to his interests).

The latest paper reprinted here [P10] reports a discovery: the soft mode. Many crystalline solids change their structure as a function of temperature, pressure, or both. If the change or transition is continuous, one can imagine a particular distortion or movement of atoms leading to the new structure. Now in a crystalline, harmonic solid, all arrangements of atoms can be described in terms of normal modes of atomic oscillations about a mean position. Thus as the point of transition is approached, that mode of oscillation which corresponds to the displacements leading to the new structure becomes soft, *i.e.* easily excited or of low frequency. This was exactly what Raman and Nedungadi observed in quartz. The particular lattice-vibration mode corresponding to the symmetry change ($\alpha \rightarrow \beta$ quartz) was seen to become softer and softer as the transition temperature $T$ is approached. In the 1960s Cochran as well as Anderson showed that the square of the soft mode frequency is proportional to $(T - T_c)$. The subject of structural transitions and soft modes saw considerable activity in the two decades 1960–1979; the pioneering work of Raman and Nedungadi was noted to be the beginning of it all.

I am thankful to Professors P. S. Narayanan, S. Ramaseshan, and C. N. R. Rao for help and advice about the selection of papers, and to Professor Ramaseshan for a conversation regarding Raman's contributions as well as style.

# Selected papers of Raman

## Contents

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[P1]

[JANUARY 15, 1920

NO. 2620, VOL. 104] 500

## Musical Drums with Harmonic Overtones.

It is well known that percussion instruments as a class give inharmonic overtones, and are thus musically defective. We find on investigation that a special type of musical drum which has long been known and used in India forms a very remarkable exception to the foregoing rule, as it gives harmonic overtones having the same relation of pitch to the fundamental tone as in stringed instruments. Five such harmonics (inclusive of the fundamental tone) can be elicited from the drumhead in this type of instrument, the first, second, and third harmonics being specially well sustained in intensity and giving a fine musical effect. The special method of construction of the drumhead which secures this result will be understood from the accompanying illustration (Fig. 1). It will be noticed (1) that the drumhead carries a symmetrical distributed load, decreasing in superficial density from the centre outwards (this appears as a dark circle in the middle of the membrane, the load consisting of a firmly adherent but flexible composition, in which the principal constituent is finely divided metallic iron); and (2) that a second membrane in the form of a ring is superimposed on the circular membrane round its margin.

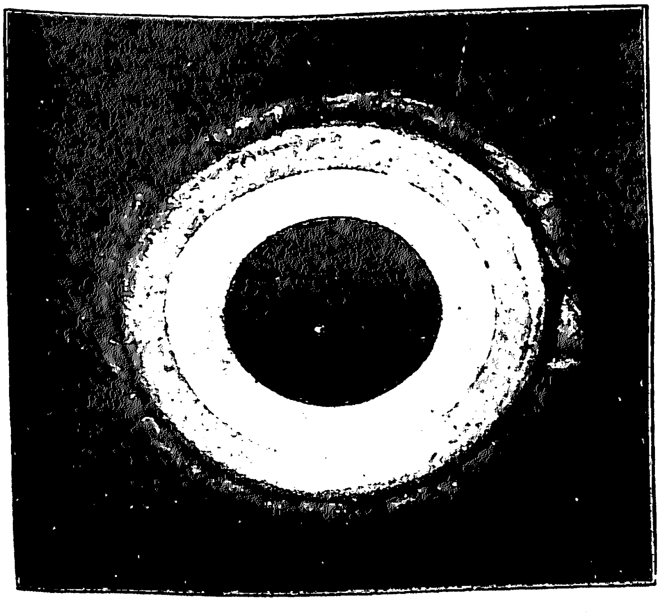

Fig. 1.—Drumhead giving harmonic overtones.

The character of the vibrations of this heterogeneous membrane which give rise to its remarkable acoustic properties have been investigated by us. It is found, as might have been expected, that the fundamental pitch and the octave are derived respectively from the modes of vibration of the membrane without any nodal lines and with one nodal diameter. The third harmonic, we find, owes its origin to the fact that the next two higher modes of vibration of the drumhead (those with two nodal diameters and with one nodal circle respectively) have identical pitch, this being a twelfth above the fundamental. There is reason to believe that the fourth and fifth harmonics similarly arise from some of the numerous more complex modes of vibration of the drumhead becoming unified in pitch in consequence of the distributed load at the centre and round the periphery of the membrane. The central load also improves the musical effect by increasing the energy of vibration, and thus prolonging the duration of the tones.

C. V. RAMAN.
SIVAKALI KUMAR.
210 Bowbazaar Street, Calcutta, India,
December 10.

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[P2]

NOVEMBER 17, 1921]

NO. 2716, VOL. 108] 367

## The Colour of the Sea.

The view has been expressed that "the much-admired dark blue of the deep sea has nothing to do with the colour of water, but is simply the blue of the sky seen by reflection" (Rayleigh's Scientific Papers," vol. 5, p. 540, and NATURE, vol. 83, p. 48, 1910). Whether this is really true is shown to be questionable by a simple mode of observation used by the present writer, in which surface-reflection is eliminated, and the other factors remain the same. The method is to view the surface of the water through a Nicol's prism, which may for convenience be mounted at one end of a tube so that it can be turned about its axis and pointed in any direction. Observing a tolerably smooth patch of water with this held in front of the eye at approximately the polarising angle with the surface of the sea, the reflection of the sky may be quenched by a suitable orientation of the Nicol. Then again, the sky-light on a clear day in certain directions is itself strongly polarised, and an observer standing with his back to the sun when it is fairly high up and viewing the sea will find the light reflected at all incidences sufficiently well polarised to enable it to be weakened or nearly suppressed by the aid of a Nicol.

Observations made in this way in the deeper waters of the Mediterranean and Red Seas showed that the colour, so far from being impoverished by suppression of sky-reflection, was wonderfully improved thereby. A similar effect was noticed, though somewhat less conspicuously, in the Arabian Sea. It was abundantly clear from the observations that the blue colour of the deep sea is a distinct phenomenon in itself, and not merely an effect due to reflected sky-light. When the surface-reflections are suppressed the hue of the water is of such fullness and saturation that the bluest sky in comparison with it seems a dull grey.

By putting a slit at one end of the tube and a grating over the Nicol in front of the eye, the spectrum of the light from the water can be examined. It was found to exhibit a concentration of energy in the region of shorter wave-lengths far more marked than with the bluest sky-light.

Even when the sky was completely overcast the blue of the water could be observed with the aid of a Nicol. It was then a deeper and fuller blue than ever, but of greatly enfeebled intensity. The altered appearance of the sea under a leaden sky must thus be attributed to the fact that the clouds screen the water from the sun's rays rather than to the incidental circumstance that they obscure the blue light of the sky.

Perhaps the most interesting effect observed was that the colour of the water (as seen with the Nicol held at the polarising angle to the surface of the water and quenching the surface-reflection) varied with the *azimuth* of observation relatively to the plane of incidence of the sun's rays on the water. When the plane of observation and the plane of incidence were the same, and the observer had his back to the sun and looked down into the water, the colour was a brilliant, but comparatively lighter, blue. As the plane of observation is swung round the colour becomes a deeper and darker blue, and at the same time decreases in intensity, until finally when the plane of observation has swung through nearly 180° the water appears very dark and of a colour approaching indigo. Both the colour and the intensity also varied with the altitude of the sun.

The dependence of the colour on the azimuth of observation cannot be explained on a simple absorption theory, and must evidently be regarded as a *diffraction* effect arising from the passage of the light through the water. Looking down into the water with a Nicol in front of the eye to cut off the surface-reflections, the track of the sun's rays could be seen entering the water and appearing by virtue of perspective to converge to a point at a considerable depth inside it. The question is: What is it that diffracts the light and makes its passage visible? An interesting possibility that should be considered in this connection is that the diffracting particles may, at least in part, be the *molecules* of the water themselves. As a rough estimate, it was thought that the tracks could be seen to a depth of 100 metres, and that the intensity of the light was about one-sixth of that of the light of the sky from the zenith. If we assume that clear water, owing to its molecular structure, is capable of scattering light eight times as strongly as dust-free air at atmospheric pressure, it is clear that the major part of the observed effect may arise in this way.

It is useful to remember that the reflecting power of water at normal incidence is quite small (only 2 per cent.), and becomes large only for very oblique reflection. It is only when the water is quite smooth and is viewed in a direction nearly parallel to the surface that the reflected sky-light overpowers the light emerging from within the water. In other cases the latter has a chance of asserting itself.

C. V. Raman.

S.S. *Narkunda*, Bombay Harbour,
September 26.

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[P3]

[February 2, 1922
No. 2727, Vol. 109] 138, 139

## Molecular Structure of Amorphous Solids.

A question of fundamental importance in the theory of the solid state is the nature of the arrangement of the ultimate particles in amorphous or vitreous bodies, of which glass is the most familiar example. Is it to be supposed that the molecules are packed together at more or less uniform distances apart, as in crystals, the orientation of individual molecules or of groups of molecules being, however, arbitrary? Or, on the other hand, is the spacing of the molecules itself irregular, the solid exhibiting in a more or less *permanent form* local fluctuations of density similar to those that arise *transitorily* in liquids owing to the movement of the molecules? The physical properties of amorphous solids, notably their softening and viscous flow below the temperature of complete fusion, would tend to support the latter view, but the possibility of a closer approximation to the crystalline state should not entirely be ruled out, especially in view of the very interesting recent work of Lord Rayleigh on the feeble double refraction exhibited by fused silica (Proc. Roy. Soc., 1920, p. 284). A good deal might be expected to depend on the nature of the material, its mode of preparation, and heat treatment. A material formed by simple fusion and re-solidification of comparatively simple molecules, such as silicon dioxide, might stand on a different footing from a material such as ordinary glass built up by chemical action and formation of complex silicates.

If the arrangement of molecules in a vitreous body were irregular, the local fluctuations of optical density would result in a strong scattering of a beam of light passing through it, the intensity of such scattering being comparable with that occurring in the liquid state at the temperature of fusion of the material (see note by the present writer in Nature of November 24 last, p. 402). On the other hand, if the arrangement of the molecules approximated to the crystalline state the scattering of light would be merely that due to the thermal movements of the molecules and would be much smaller. As a matter of fact, glasses exhibit a very strong scattering of light, some 300 to 500 times as strong as in dust-free air, the Tyndall cone being of a beautiful sky-blue colour and nearly, but not quite, completely polarised when viewed in a transverse direction. (Some glasses exhibit a green, yellow, or pink fluorescence when a beam of sunlight is focussed within them, and cannot be used for the present purpose; the fluorescence, even when very feeble, can be detected by the difference in colour of the two images of the Tyndall cone seen through a double-image prism.) Rayleigh, who observed the light-scattering in glass, attributed it to inclusions, some of which he assumed must be comparable in size with the wave-length (Proc. Roy. Soc., 1919, p. 476). The closest scrutiny through the microscope under powerful dark-ground illumination fails, however, to indicate the presence of any such inclusions, and it seems more reasonable to assume, in view of the foregoing remarks, that the scattering is really molecular. Its magnitude is of the order that might be expected on the basis of a non-uniform distribution of the molecules.

Further observations with specially prepared glasses and with fused silica would be of great interest to investigate the influence of the chemical constitution and heat treatment on the molecular *texture* of the solid.
C. V. RAMAN.

210 Bowbazaar Street, Calcutta, December 29.

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[P4]

APRIL 23, 1927]

No. 2999, VOL. 119] 601

## X-ray Diffraction in Liquids.

IN order to find experimental support for the theory of X-ray diffraction in liquids put forward some three years ago by C. V. Raman and K. R. Ramanathan (*Proc. Indian Association for the Cultivation of Science*, vol. 8, p. 127, 1923), extensive studies have been undertaken in the authors' laboratory of the phenomena observed when a pencil of monochromatic X-rays passes through a layer of fluid, particularly with the view of determining how the effects are influenced by the physical condition and the chemical nature of the substance under investigation. The photographs here reproduced (Fig. 1, *a* and *b*) were obtained in the course of work on this line by one of us (C. M. Sogani) and represent the X-ray liquid-haloes of hexane and cyclo-hexane respectively. The fluids were contained in cells with very thin walls of mica, and the *K*-radiation of copper from a Shearer X-ray tube was used.

The differences between the two patterns are sufficiently striking; cyclo-hexane shows a bright and sharply defined halo with a very clear dark space within, while hexane, on the other hand, shows a less intense and relatively diffuse halo, the inner margin of which is not sharply terminated but extends almost up to the direction of the incident rays. These differences indicate very clearly the effect of the geometrical form of the molecules on the X-ray scattering by a liquid. From an X-ray point of view, cyclo-hexane consisting of ring-formed—though arbitrarily orientated—molecules has a nearly homogeneous structure, while on the other hand the elongated shape and varying orientations of the molecules in hexane cause it to be much less homogeneous in X-ray scattering. This explanation is supported by the observation that the diffraction halo of benzene resembles very closely that of cyclo-hexane.

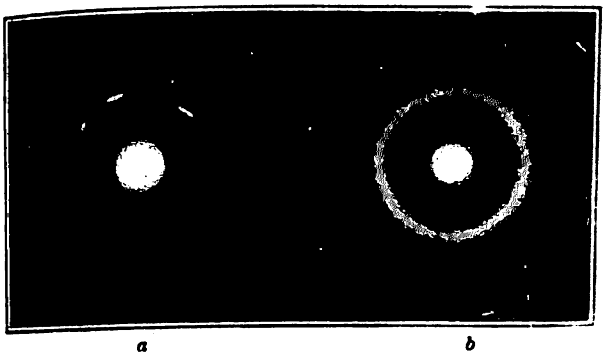

FIG. 1.—X-ray diffraction haloes of liquids. *a*, Hexane; *b*, cyclo-hexane.

It is very interesting to contrast these facts with the optical behaviour of the three liquids with regard to the scattering of ordinary light. Optically, hexane and cyclo-hexane are far more nearly similar to each other, and differ strikingly from benzene, the depolarisation of the scattered light being small for hexane and cyclo-hexane and relatively large for benzene. Here, evidently, the geometrical form of the molecule is of much less importance than its chemical character.

Further studies of the liquid-haloes for various organic substances of the aromatic and aliphatic series, and specially with the long-chain compounds, are in progress.
C. V. RAMAN.
C. M. SOGANI.

210 Bowbazar Street,
Calcutta, India, Feb. 10.

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[P5]

[JULY 30, 1927

No. 3013, VOL. 120] 158

## Optical Behaviour of Protein Solutions.

A VERY remarkable increase in light-scattering power is exhibited by gelatine solutions when the hydrogen-ion concentration approaches the value (about $pH = 5$) corresponding to the iso-electric point. This effect, which appears to have been known for some time, has been recently studied in detail by Kraemer and his co-workers, who give interesting curves showing the manner in which the Tyndall effect varies with $pH$ and temperature ("Colloid Symposium Monograph," vol. 4, and *Journal of Physical Chemistry*, May 1927).

The phenomena are scarcely intelligible on the view commonly adopted that the Tyndall effect in a colloidal solution is simply proportional to the number of scattering particles of the same kind present in it. Their explanation becomes clearer if we apply to colloidal solutions the general thermodynamic theory of light scattering, in which the Tyndall effect is regarded as due to local fluctuations of optical density in the medium. According to the latter theory, the scattering power of a colloidal solution would be connected with the osmotic pressure $P$ of the particles, by the relation

$$\frac{\pi^2 RT}{2N\lambda^4}\,\frac{k(\partial\epsilon/\partial k)^2\rho/m}{\partial P/\partial k}, \quad . \quad . \quad . \quad (1)$$

where $k$ is the concentration of the dispersed material, $\epsilon$ is the optical dielectric constant of the solution and $\rho/m$ is practically unity for a dilute solution. It is well known from the work of Jacques Loeb that the osmotic pressure of a gelatine solution alters in a notable manner with $pH$, becoming very small at the iso-electric point. Equation (1) then enables us to see at once why the Tyndall effect becomes very large under the same conditions.

A detailed discussion of colloidal optics on the basis of the thermodynamic theory of light scattering will be published in the *Indian Journal of Physics*.

C. V. RAMAN.

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[P6]

APRIL 21, 1928]

No. 3051, VOL. 121] 619

## A Change of Wave-length in Light Scattering.

FURTHER observations by Mr. Krishnan and myself on the new kind of light-scattering discovered by us have been made and have led to some very surprising and interesting results.

In order to convince ourselves that the secondary radiation observed by us was a true scattering and not a fluorescence, we proceeded to examine the effect in greater detail. The principal difficulty in observing the effect with gases and vapours was its excessive feebleness. In the case of substances of sufficient light-scattering power, this difficulty was overcome by using an enclosed bulb and heating it up so as to secure an adequate density of vapour. Using a blue-violet filter in the track of the incident light, and a complementary green-yellow filter in front of the observer's eye, the modified scattered radiation was observed with a number of organic vapours, and it was even possible to determine its state of polarisation. It was found that in certain cases, for example, pentane, it was strongly polarised, while in others, as for example naphthalene, it was only feebly so, the behaviour being parallel to that observed in the liquid state. Liquid carbon dioxide in a steel observation vessel was studied, and exhibited the modified scattering to a notable extent. When a cloud was formed within the vessel by expansion, the modified scattering brightened up at the same time as the ordinary or classical scattering. The conclusion is thus reached that the radiations of altered wave-length from neighbouring molecules are coherent with each other.

A greater surprise was provided by the spectroscopic observations. Using sunlight with a blue filter as the illuminant, the modified scattered radiation was readily detected by the appearance in the spectrum of the scattered light of radiations absent from the incident light. With a suitably chosen filter in the incident light, the classical and modified scatterings appeared as separate regions in the spectrum separated by a dark region. This encouraged us to use a mercury arc as the source of light, all radiations of longer wave-length than 4358 A. being cut out by a filter. The scattered radiations when examined with a spectroscope showed some sharp bright lines additional to those present in the incident light, their wave-length being longer than 4358 A.; at least two such lines were prominent and appeared to be accompanied by some fainter lines, and in addition a continuous spectrum. The relation of frequencies between the new lines and those present in the incident light is being investigated by photographing and measuring the spectra. The preliminary visual observations appear to indicate that the position of the principal modified lines is the same for all substances, though their intensity and that of the continuous spectrum does vary with their chemical nature.

C. V. RAMAN.

210 Bowbazar Street,
Calcutta, Mar. 8.

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[P7]

MAY 5, 1928]

No. 3053, VOL. 121] 711

## The Optical Analogue of the Compton Effect.

THE presence in the light scattered by fluids, of wave-lengths different from those present in the incident light, is shown very clearly by the accompanying photographs (Fig. 1). In the illustration (1) represents the spectrum of the light from a quartz mercury vapour lamp, from which all wave-lengths greater than that of the indigo line have been filtered out. This line (4358 A.) is marked D in the spectrogram, and C is the group of lines 4047, 4078, and 4109 A. Spectrogram (2) shows the spectrum of the scattered light, the fluid used being toluene in this case. It will be seen that besides the lines present in the incident spectrum, there are several other lines present in the

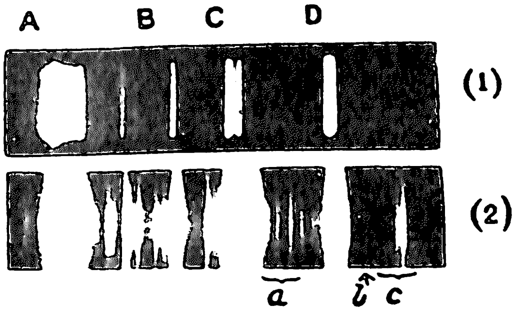

FIG. 1.—(1) Spectrum of incident light; (2) spectrum of scattered light.

scattered spectrum. These are marked *a, b, c* in the figure, and in addition there is seen visually another group of lines which is of still greater wave-length and lies in a region outside that photographed. When a suitable filter was put in the incident light to cut off the 4358 line, this latter group also disappeared, showing that it derived its origin from the 4358 line in the incident radiation. Similarly, the group marked *c* in spectrogram (2) disappeared when the group of lines 4047, 4078 and 4109 was filtered out from the incident radiation by quinine solution, while the group due to 4358 A. continued to be seen. Thus the analogy with the Compton effect becomes clear, except that we are dealing with shifts of wave-length far larger than those met with in the X-ray region.

As a tentative explanation of the new spectral lines thus produced by light-scattering, it may be assumed that an incident quantum of radiation may be scattered by the molecules of a fluid either as a whole or in part, in the former case giving the original wave-length, and in the latter case an increased wave-length. This explanation is supported by the fact that the diminution in frequency is of the same order of magnitude as the frequency of the molecular infra-red absorption line. Further, it is found that the shift of wave-length is not quite the same for different molecules, and this supports the explanation suggested.

Careful measurements of wave-length now being made should settle this point definitely at an early date.

C. V. RAMAN.
K. S. KRISHNAN.

210 Bowbazar Street,
Calcutta, Mar. 22.

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[P8]

[SEPTEMBER 14, 1929
No. 3124, VOL. 124] 412

## Anomalous Diamagnetism.

IN a letter in NATURE of June 22 (p. 945), reference was made to the Ehrenfest hypothesis which ascribes the high diamagnetic susceptibility of bismuth to the existence of closed electron orbits of larger than atomic dimensions in association with the crystal lattice.

It may be pointed out that the existence of such orbits appears to furnish a very natural explanation of a variety of phenomena which up to now have been obscure. In the first place, the extremely pronounced diamagnetic anisotropy characteristic of bismuth (and also of graphite) becomes immediately intelligible as a consequence of the specific orientation of the assumed electronic circulations within the crystal lattice. Further, the large Hall effects and changes of electrical resistance exhibited by bismuth and graphite when placed in a magnetic field become comprehensible, since the electronic circulations would be modified by the field, and result in corresponding modifications of the flow of electricity through the substance under a simultaneously impressed electromotive force. There would be every reason to expect, as is indeed the case, that the magnitudes of the Hall effect and the change of resistance would depend on the orientation of the crystal in the magnetic field and the direction of flow of electricity through it.

Then again, with rise of temperature and consequent thermal derangements of the lattice, the postulated electronic circulations would tend to disappear and give place to chaotic electronic movements. The diamagnetic susceptibility would then diminish towards its normal value for a non-crystalline condition of the substance, and corresponding changes would occur in the coefficients of the Hall effect and magnetic variation of electrical resistance. That liquid metals do not, so far as I am aware, exhibit a measurable Hall effect is significant in this connexion.

In close analogy with the influence of temperature, and presumably to be explained on very similar lines, is the remarkable fact that the anomalous diamagnetism of bismuth and of graphite tends to diminish or disappear when the substances are reduced to a colloidal condition.

Finally, it may be remarked that the dimensions of the crystal lattice cannot be uninfluenced by the existence of such regular electronic circulations within it or by modifications produced in them by an external agency. That the magnetostriction of bismuth in strong fields discovered by Dr. Kapitza (NATURE, July 13, p. 53) is connected with the anomalous diamagnetism of the substance admits of little doubt. The notable increase in magnetostriction at low temperatures observed by him appears to fit in very well with the Ehrenfest hypothesis.

C. V. RAMAN

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[P9]

FEB, 5, 1938

No. 3562, VOL. 141 242 243

## Light Scattering and Fluid Viscosity

ACCORDING to well-known hydrodynamical theory[1], plane waves of sound propagated through a viscous liquid suffer a diminution of amplitude in the ratio $1/e$ in traversing a number of wave-lengths given by the quantity $3C\lambda/8\pi^2\nu$, where $C$ is the velocity of sound, $\lambda$ is the wave-length of sound and $\nu$ is the kinematic viscosity. Taking $\lambda = 4358$ A., this number for various common liquids which are fairly mobile at room temperature ranges from about 3 in the case of butyl alcohol to about 30 in the case of carbon disulphide. For phenol at 25° C., the number is less than 1, and for glycerine, it is a small fraction of unity. A consideration of these numbers shows that the theories due to Einstein[2] and L. Brillouin[3], which regard the diffusion of light occurring in liquids as due to the reflection of light by regular and infinitely extended trains of sound-waves present in them, can only possess partial validity for ordinary liquids, and must break down completely in the case of very viscous ones. In an earlier note in NATURE[4], we reported studies of the Fabry-Perot patterns of scattered light with a series of liquids, which showed clearly that the Doppler-shifted components in the spectrum of scattered light fell off in intensity relatively to the undisplaced components, with increasing viscosity of the liquid.

We have now to report some further results which illustrate in a striking way the part played by fluid viscosity in the diffusion of light by liquids. As mentioned in our previous note, the light scattered by liquid phenol at ordinary temperatures gives a Fabry-Perot pattern which is scarcely distinguishable from that of the incident light. When, however, the temperature of the liquid is raised, the viscosity falls off rapidly, and the number $3C\lambda/8\pi^2\nu$ assumes a value which is many times greater than at room temperature. Simultaneously, as can be seen from Fig. 1, the character of the Fabry-Perot pattern alters, and the Doppler-shifted components come increasingly into evidence; at 70° C. they are just as prominent as in ordinary inviscid liquids. The influence of temperature revealed by these studies for the case of the very viscous phenol is to be clearly distinguished from the broadening of the Doppler components with rise of temperature reported by us in an earlier note[5] for the case of carbon tetrachloride.

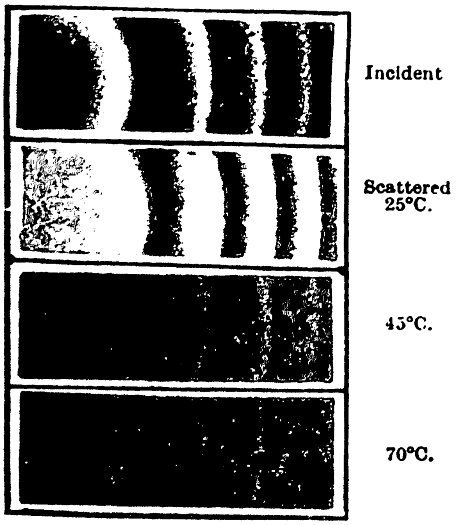

Fig. 1.

INFLUENCE OF TEMPERATURE ON THE SCATTERING BY PHENOL.

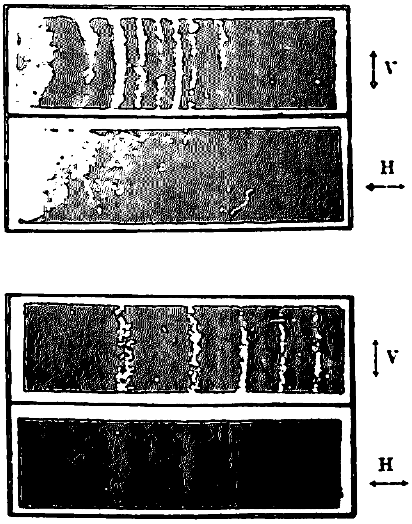

Fig. 2.

STATE OF POLARIZATION; ABOVE, TOLUENE; BELOW, PHENOL.

The four patterns reproduced in Fig. 2 show the remarkable difference in the state of polarization of the Fabry-Perot patterns of transversely scattered light for an inviscid liquid such as toluene and a viscous one such as phenol at room temperature. In the former case, only a continuous radiation is to be observed in the horizontal component; in other words, both the displaced and the undisplaced components in the pattern are sensibly completely polarized with the vibrations vertical. In the case of phenol, however, the undisplaced radiation is evidently partially polarized, as it appears both in the vertical and the horizontal vibrations; a partial polarization of the continuous radiation is also noticeable.

C. V. RAMAN.
B. V. RAGHAVENDRA RAO.

Department of Physics,
Indian Institute of Science,
Bangalore.
Dec. 29.

Lamb, "Hydrodynamics", fifth edition, p. 613.
Einstein, A., *Ann. Phys.*, **33**, 1275 (1910).
Brillouin, L., *Ann. Phys.*, **17**, 88 (1922).
NATURE, **139**, 585 (April 3, 1937).
NATURE, **135**, 761 (May 4, 1935).

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[P10]

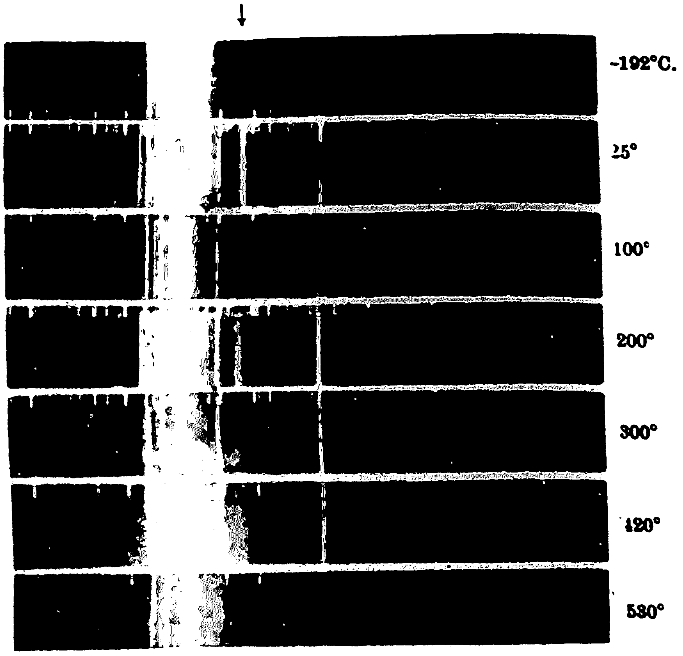
JAN. 27, 1940
No. 3665, VOL. 145 147

# The α-β Transformation of Quartz

As is well known, the ordinary form of quartz which has trigonal symmetry changes over reversibly to another form which has hexagonal symmetry at a temperature of 575° C. Though the transformation does not involve any radical reorganization of the internal architecture[1] of the crystal and takes place at a sharply defined temperature, it is nevertheless preceded over a considerable range of temperature (200°–575°) by a progressive change in the physical properties of 'low' quartz which prepares the way for a further sudden change, when the transition to 'high' quartz actually takes place. The thermal expansion coefficients, for example, gradually increase over this range of temperature, becoming practically infinite at the transition point and then suddenly dropping to small negative values[2]. Young's moduli in the same temperature range fall to rather low values at the transition point and then rise sharply to high figures[3]. The piezo-electric activity also undergoes notable changes[4,5].

In the hope of obtaining an insight into these remarkable phenomena, a careful study has been made of the spectrum of monochromatic light scattered in a quartz crystal at a series of temperatures ranging from that of liquid air to nearly the transition point. Significant changes are observed which are illustrated in the accompanying illustration, reproducing part of the spectrum excited by the 4358 A. radiation of the mercury arc. A fully exposed spectrum at room temperature indicates fourteen different normal modes of vibration of the crystal. At liquid air temperature, the three most intense lines correspond to the frequency shifts 132, 220 and 468 cm.$^{-1}$ and are all about equally sharp. As the crystal is heated over the temperature range 200°–530°, notable changes occur. The 220 cm.$^{-1}$ line (marked with an arrow in the reproduction) behaves in an exceptional way, spreading out greatly towards the exciting line and becoming a weak diffuse band as the transition temperature is approached. On the other hand, the other intense lines having both larger and smaller frequency shifts continue to be easily visible, though appreciably broadened and displaced.

The behaviour of the 220 cm.$^{-1}$ line clearly indicates that the binding forces which determine the frequency of the corresponding mode of vibration of the crystal lattices diminish rapidly with rising temperature. It appears therefore reasonable to infer that the increasing excitation of this particular mode of vibration with rising temperature and the deformations of the atomic arrangement resulting therefrom are in a special measure responsible for the remarkable changes in the properties of the crystal already mentioned, as well as for inducing the transformation from the α to the β form.



LIGHT SCATTERING IN QUARTZ.

C. V. RAMAN.
T. M. K. NEDUNGADI.

Department of Physics,
Indian Institute of Science,
Bangalore.
Dec. 11.

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[1] Bragg and Gibbs, *Proc. Roy. Soc.*, A, **109**, 405 (1925).
[2] Jay, *Proc. Roy. Soc.*, A, **142**, 237 (1933).
[3] Perrier and Mandrot, *C.R.*, **175**, 622 (1922).
[4] Osterberg and Cookson, *J. Frank. Inst.*, **220**, 361 (1935).
[5] Pitt and Mckinley, *Canad. J. Res.*, A, **14**, 58 (1936).

[P11] Journal
of the
Optical Society of America
and
Review of Scientific Instruments

Vol. 15 OCTOBER, 1927 Number 4

# THE SCATTERING OF LIGHT IN AMORPHOUS SOLIDS

BY C. V. RAMAN

## 1. INTRODUCTION

Recent investigations have shown that when light traverses a dust-free liquid, an observable fraction of the energy is laterally scattered and that this effect is due to the local fluctuations of density and to the random orientations of the molecules which cause the fluid to be optically inhomogeneous.[1] In the case of a mixture of liquids, we have in addition a scattering due to the local fluctuations of composition which cause corresponding local variations of refractive index. Since the transverse scatterings due to density and composition fluctuations are fully polarized, while that due to the random orientation of the molecules is almost entirely unpolarized, the resultant scattering in a fluid is usually only partially polarized. When the temperature of a liquid is lowered, its compressibility usually diminishes and with it also the local fluctuations of density. Thus we may expect that when the liquid is cooled to such an extent that it passes into the amorphous solid condition, the density scattering would become very small. On the other hand, in liquid mixtures, the local fluctuations of *composition* usually tend to increase rather than to diminish with fall of temperature. Thus they should certainly tend to persist or even increase when the mixture congeals into an amorphous solid. The effect due to random molecular orientations would certainly remain in the amorphous solid condition. Thus, we may anticipate that an amorphous solid such as glass consisting of a mixture of anisotropic molecules would exhibit when light traverses it, a partially-polarized internal scattering or opalescence of an order of intensity not greatly inferior to that ordinarily observed in liquids or liquid mixtures.

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[1] C. V. Raman, Molecular Diffraction of Light, Calcutta University Press, 1922.

Printed by permission from *J. Opt. Soc. Am.*, 1927, **15**, 185–189.

An internal scattering of light in common glasses and also in optical glasses has actually been noticed.[2] Its nature has been a subject of debate,[3] and owing to our insufficient knowledge of the amorphous state, is not fully understood at present. Judging, however, from such observations as are available, it is the opinion of the writer, that the effect observed in optical glasses is a true molecular scattering arising from local fluctuations of composition and of molecular orientation, being thus of the same general nature as the opalescence observed in binary liquid mixtures such as phenol-water, carbon disulphide-methyl alcohol and so on. In support of this view, it is proposed in this paper to give the results of the study of the light-scattering in a series of 14 different optical glasses manufactured by the firm of Schott in Jena.[4] If the light-scattering in glass were due to accidental inclusions or incipient crystallizations occurring within it as has been suggested by some writers, we should expect the intensity of the scattered light to show large and arbitrary variations depending on the circumstances of the particular melting from which the specimen was taken. On the other hand, if the phenomenon has a true molecular origin, we should expect to find the intensity of scattering to be definitely correlated with the refractivity and chemical constitution of the glass.

TABLE 1. *List of classes examined.*

| | Melting | Type | Composition | Refractive index $\mu$ | $\nu = 1/\omega$ |
|---|---|---|---|---|---|
| No. 1 | 18165 | 0.6781 | Fluor crown | 1.4933 | 69.9 |
| 2 | 4927 | U. V. 3199 | U. V. crown | 1.5035 | 64.4 |
| 3 | 15189 | 0.144 | Borosilicate crown | 1.5100 | 64 |
| 4 | 16564 | 0.3832 | Prism crown | 1.5163 | 64 |
| 5 | 15065 | 0.3453 | Silicate crown | 1.5191 | 60.4 |
| 6 | 16740 | 0.3439 | Telescopic flint | 1.5286 | 51.6 |
| 7 | 16397 | 0.7550 | Baryta light flint | 1.5694 | 56 |
| 8 | 14657 | 0.211 | Densest Borosilicate crown | 1.5726 | 57.5 |
| 9 | 17538 | 0.340 | Ordinary Light flint | 1.5774 | 41.4 |
| 10 | 18023 | 0.1266 | Baryta light flint | 1.6042 | 43.8 |
| 11 | 11095 | 0.103 | Ordinary flint | 1.6202 | 36.2 |
| 12 | 10259 | 0.748 | Baryta flint | 1.6235 | 39.1 |
| 13 | 8988 | 0.102 | Dense flint | 1.6489 | 33.8 |
| 14 | 16889 | 0.198 | Densest flint | 1.7782 | 26.5 |

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[2] Lord Rayleigh, Proc. Roy. Soc., *95*, p. 476, 1919, and C. V. Raman, "Molecular Diffraction of Light," p. 85.

[3] R. Gans, Ann. der Phys., *77*, p. 317; 1925.

[4] The specimens were presented by Messrs. Schott to Prof. P. N. Ghosh, who kindly placed them at the writer's disposal for the work.

## 2. THE SPECIMENS EXAMINED

Table 1 gives a list of the glasses examined and their description as furnished by the manufacturers, arranged in order of increasing refractive index. The samples were furnished in the form of slabs 7 cm×7 cm×2 cm, with one pair of end-faces polished. For the purpose of the observation of light-scattering, the slabs were immersed in a trough containing benzene and a beam of sunlight focused by a lens was admitted through a side-face, the necessity of polishing the latter being thus avoided. The track of the beam as seen through the end-faces was perfectly uniform and appeared of a beautiful sky-blue color.

## 3. EXPERIMENTAL RESULTS

When the scattered light was viewed through a double-image prism held so that the direction of vibrations in the two images seen were respectively perpendicular and parallel to the direction of the beam traversing the glass, it was seen that these were of very different intensities, showing that the scattered light was strongly but not completely polarized. The color of the stronger image was always a sky-blue. The color of the fainter image in the ordinary flint glasses was blue, but in the other specimens varied very considerably. The total intensity of the scattered light in the glasses was determined by comparison with that of the track in a bulb containing dust-free benzene immersed in the same trough as the block of glass under examination and traversed by the same beam of light. A rotating-sector photometer was used for the purpose. The ratio of the intensities in the parallel and perpendicular components of vibration in the laterally scattered light was also determined with the help of a double-image prism and nicol (Cornu's method) in the usual way. The measurements give us the ratio of the intensity of the faint image to the bright image seen through the double image prism, and this expresses the degree of depolarization of the scattered light.

The results of the work are gathered together in Table 2.

## 4. DISCUSSION OF RESULTS

From a scrutiny of the figures in Table 2, several interesting facts emerge. In the first place, it will be seen that the crown glasses show uniformly a smaller intensity of light-scattering than the other varieties of glass. Both the ordinary flints and the baryta flints scatter light strongly, the latter more so than ordinary flints of equal refractive index. It will be seen also that considering each species of glass separately, there is a progressive increase of the intensity of light-scattering with increasing refractive-index. The colors shown by the fainter components of the scattered light in the first four glasses in Table 2 are obviously due to a species of weak fluorescence, probably of the same kind as has been met with in investigations on light-scattering in liquids.[5] This fluorescence being unpolarized, the large values of the depolarization found in the case of these glasses stands self-explained. If the fluorescent light had been excluded by the introduction of suitable color-filters, the depolarization for these glasses would have been much smaller. It is interesting to notice that in glasses Nos. 6, 7, and 8, we have a low value for the depolarization in spite of the obvious presence of a weak fluorescence; this is obviously due to the greater intensity of polarized scattering appearing in the last column of Table 2 of these glasses.

TABLE 2. *Experimental results.*

| | Glass | Refractive index | Color of bright component | Color of faint component | Depolarization | Intensity relative to benzene = 1 |
|---|---|---|---|---|---|---|
| No. 1 | Fluor crown | 1.4933 | Blue | Yellow | 0.180 | 0.18 |
| 2 | U. V. crown | 1.5035 | “ | Pink | 0.158 | 0.12 |
| 3 | Borosilicate crown | 1.5100 | “ | “ | 0.295 | 0.11 |
| 4 | Prism crown | 1.5163 | “ | “ | 0.285 | 0.14 |
| 5 | Silicate crown | 1.5191 | “ | Blue | 0.123 | 0.18 |
| 6 | Telescopic flint | 1.5286 | “ | Purple | 0.068 | 0.40 |
| 7 | Baryta light flint | 1.5694 | “ | “ | 0.053 | 0.44 |
| 8 | Densest borosilicate crown | 1.5726 | “ | “ | 0.045 | 0.37 |
| 9 | Ordinary light flint | 1.5774 | “ | Indigo-blue | 0.067 | 0.30 |
| 10 | Baryta light flint | 1.6042 | “ | Indigo | 0.067 | 0.41 |
| 11 | Ordinary flint | 1.6202 | “ | Blue | 0.079 | 0.42 |
| 12 | Baryta flint | 1.6235 | “ | Indigo-blue | 0.085 | 0.57 |
| 13 | Dense flint | 1.6489 | “ | Blue | 0.062 | 0.52 |
| 14 | Densest flint | 1.7782 | “ | “ | 0.065 | 0.63 |

The several regularities to which attention has been drawn above, particularly the fact that the intensity of scattering is very clearly a function of the refractive-index and chemical composition of the glass, render it extremely improbable that the effect can arise from accidental inclusions or imperfections in the structure of the glasses. It is, in fact, clear from the data that the effect arises from the ultimate molecular structure of glass.

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[5] K. S. Krishnan, Phil. Mag., *50*, p. 697; 1925.

It would be very interesting to study the light-scattering in amorphous solids having a relatively simple chemical constitution, e.g., transparent quartz-glass. Experiments on the scattering of light in liquids which can first be rendered dust-free and then supercooled into the amorphous solid state may also be expected to furnish important information. Further work on these lines is in progress.

210 BOWBAZAR STREET,
CALCUTTA, INDIA,
MAY 11, 1927.

[P12]

## 24

# A New Radiation[1]

BY

PROF. C. V. RAMAN, F.R.S.

(Plate XII).

## 1. *Introduction.*

I propose this evening to speak to you on a new kind of radiation or light-emission from atoms and molecules. To make the significance of the discovery clear, I propose to place before you the history of the investigations made at Calcutta which led up to it. Before doing so, however, a few preliminary remarks regarding radiation from atoms and molecules will not be out of place.

Various ways are known to the physicist by which atoms or molecules may be caused to emit light, as for instance, heating a substance or bombarding it with a stream of electrons. The light thus emitted is usually characteristic of the atoms or molecules and is referred to as *primary* radiation. It is also possible to induce radiation from atoms and molecules by illuminating them strongly. Such light-emission is referred to as *secondary* radiation. The familiar diffusion of light by rough surfaces may be cited as an example of secondary radiation, but strictly speaking, it hardly deserves the name, being an effect occuring at the boundaries between media of different refractive indices and not a true volume-effect in which all the atoms and molecules of the substance

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[1] Inaugural Address delivered to the South Indian Science Association on Friday, the 16th March, 1928, at Bangalore.

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PLATE XII

**Fig. 1**

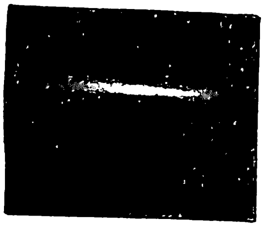

**Fig. 2.**

Polarisation
of
Scattering.
Fig 1 Unmodified
Fig 2 Modified

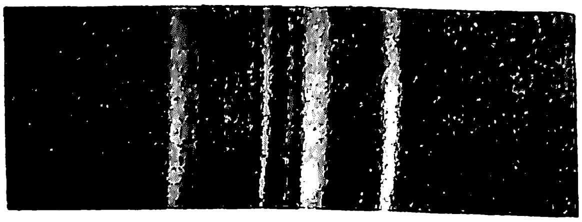

Fig 3 (1)
Incident Spectrum

Fig 3 (2)
Scattered Spectrum

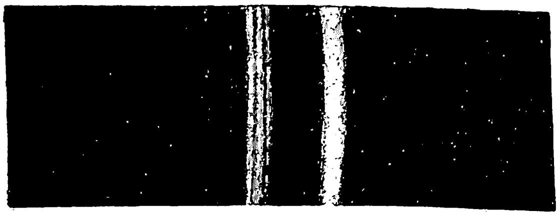

Fig 4 (1)
Incident Spectrum

Fig 4 (2)
Scattered Spectrum

take part. The first case discovered of secondary radiation really worthy of the name was the phenomenon of fluorescence whose laws were elucidated by the investigations of Sir George Stokes. This is a familiar effect which is exhibited in a very conspicous manner in the visible region of the spectrum by various organic dye-stuffs. I have here a bottle of water in which an extremely small quantity of fluorescein is dissolved. You notice that when placed in the beam of light from the lantern, it shines with a vivid green light, and that the colour of the emission is not altered, though its brightness is changed, by placing filters of various colours between the bottle and the lantern. A violet filter excites the green fluorescence strongly, while a red filter has but little effect.

Another kind of secondary radiation whose existence has been experimentally recognized more recently is the scattering of light by atoms and molecules. It is this scattering that gives us the light of the sky, the blue colour of the deep sea and the delicate opalescence of large masses of clear ice. I have here a large bottle of a very clear and transparent liquid, toluene, which as you notice contains hardly any dust-particles, but the track of the beam from the lantern passing through it is visible as a brilliant blue cone of light. This internal opalescence continues to be visible even after the most careful purification of the liquid by repeated distillation in vacuo. A similar opalescence is shown, though much less brightly, by dust-free gases and vapours, and also by solids. A large clear block of ice shows a blue colour in the track of the beam when sunlight passes through it. The blue opalescence of blocks of clear optical glass is also readily demonstrable. The molecular scattering of light is thus a phenomenon common to all states of matter.

During the past seven years, the scattering of light in transparent media has been the subject of intensive experimental and theoretical investigation at Calcutta, and it is the researches made on this subject that have led to the discovery which I shall lay before you this evening. One important outcome of our researches has been to show that while light-scattering is in one sense a molecular phenomenon, in another sense it is a bulk-effect having a thermal origin. It is the thermal agitation of the molecules which causes them to be distributed and orientated in space with incomplete regularity, and it is the local fluctuations in the properties of the medium thus arising which give rise to optical heterogeneity and consequent diffusion of light. The subject of light-scattering is thus a meeting ground for thermodynamics, molecular physics and the wave-theory of radiation. That the combination of theories in such diverse fields of physics gives us predictions which have been experimentally verified, is one of the triumphs of modern physics.

## 2. *A New Phenomenon.*

While the quantitative investigations made at Calcutta have in the main substantiated the thermodynamic-wave-optical theory of light-scattering, indications appeared even in our earliest studies of a new phenomenon which refused to fit in with our pre-conceived notions. Thus, in some observations made by me[1] with the assistance of Mr. Seshagiri Rao in December, 1921, it was found that the depolarisation of the light transversely scattered by distilled water measured with a double-image prism and Nicol increased very markedly when a violet filter was placed in the path of the incident light. More careful investigations made with dust-free liquids[2] in 1922, confirmed this effect and showed it to exist also in methyl and ethyl alcohols, and to a lesser degree in ether. It was also noticed that the colours of the scattered light from the different liquids studied did not match

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[1] "Molecular Diffraction of Light," Calcutta University Press, February, 1922.

[2] C. V. Raman and K. S. Rao, Phil. Mag., Vol. 46, p. 633, 1923.

perfectly. An important advance was made when Dr. Ramanathan[1] working at Calcutta in the summer of 1923, investigated the phenomenon more closely and discovered that it was not a true dependance of the depolarisation on the wave-length of the scattering radiation but was due to the presence in the scattered light of what he described as "a trace of fluorescence." This was shown by the fact that the measured depolarisation depended on whether the blue filter used was placed in the path of the incident beam or of the scattered light, being smaller in the latter case. Accepting the explanation of the effect as "weak fluorescence," it naturally became important to discover whether it was due to some impurity present in the substance. Dr. Ramanathan tested this by careful chemical purification followed by repeated slow distillation of the liquid at the temperature of melting ice. He found that the effect persisted undiminished.

The investigation of this species of " weak fluorescence " has ever since 1923 been on our programme of research at Calcutta. Krishnan,[2] who investigated 60 liquids for light-scattering in the spring and summer of 1924, made systematic studies of the phenomenon, and found that it was shown markedly by water, ether, all the monohydric alcohols and a few other compounds. He pointed out that the liquids which exhibit the effect have certain family relationships amongst themselves, and that they are also substances whose molecules are known to be polar. The chemical importance of the subject led to Mr. S. Venkateswaran attempting to make a fuller study of it in the summer of 1925, but without any special success. The research was discontinued at the time but was resumed by him later in the current year (January, 1928). The remarkable observation was made that the visible radiation which is excited

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[1] K. R. Ramanathan, Proc. Ind. Assoc. Cultn. Science, Vol. VIII, p. 190, 1923.

[2] K. S. Krishnan, Phil. Mag., Vol. L, p. 697, 1925.

in pure dry glycerine by ultra-violet radiation (sunlight filtered through Corning glass G. 586) *is strongly polarised.*

The possibility of a similar effect in gases and vapours was also borne in mind and repeatedly looked for by the workers at Calcutta. The feebleness of the scattering in gases and vapours, and the infructuousness of the earlier efforts in this direction, however discouraged progress.

### 3. *Its Universality.*

Though the phenomenon was described in the paper of Dr. Ramanathan and Mr. Krishnan as a "feeble fluorescence," the impression left on my mind at the time was that we had here an entirely new type of secondary radiation distinct from what is usually described as fluorescence. The publication of the idea was however discouraged by the belief then entertained that only a few liquids exhibited the effect and by the supposition that it was unpolarised in the same way as ordinary fluorescence in liquids. Indeed, a chemical critic might even have asserted that the effect was in each case due to a trace of dissolved fluorescent impurity present in the substance which our efforts at purification had failed to remove. Early this year, however, a powerful impetus to further research was provided when I conceived the idea that the effect was some kind of optical analogue to the type of X-ray scattering discovered by Prof. Compton, for which he recently received the Nobel Prize in Physics. I immediately undertook an experimental re-examination of the subject in collaboration with Mr. K.S. Krishnan and this has proved very fruitful in results. The first step taken in the research was to find whether the effect is shown by all liquids. The method of investigation was to use a powerful beam of sunlight from a heliostat concentrated by a 7″ telescope objective combined with a short focus lens. This was passed through a blue-violet filter and then through the liquid under examination contained in an evacuated bulb and purified by repeated distillation in vacuo. A second filter of green glass was used which was complementary in colour to the blue-violet filter. If it were placed in the track of the incident light, all illumination disappears, while, if it be placed between the bulb and the observer's eye, the opalescent track within the liquid continued to be visible, though less brightly. All the liquids examined (and they were some 80 in number) showed the effect in a striking manner. There was therefore no longer any doubt that the phenomenon was universal in character; with the bulb of toluene on the lantern, you see that the effect is readily demonstrable. The cone of light vanishes when I place the violet and green filters together, but it appears when I transfer the latter to a place between my audience and the observation bulb.

Now the test with the complementary filters is precisely that ordinarily used for detecting fluorescence and indeed was first suggested by Stokes in his investigations on the subject. You may therefore rightly ask me the question how does this phenomenon differ from fluorescence? The answer to the question is, firstly, that it is of an entirely different order of intensity. A more satisfactory proof was however forthcoming when Mr. Krishnan and myself examined the polarisation of this new type of radiation and found that it was nearly as strong as that of the ordinary light scattering in many cases, and is thus quite distinct from ordinary fluorescence which is usually unpolarised.

This is shown for the case of toluene in Figs. 1 and 2 in Plate XII. Fig. 1 is a photograph of the scattering by toluene of sunlight filtered through a blue-violet glass. It was taken through a double-image prism of iceland spar with an exposure of 3 seconds. Fig. 2 is a picture with an additional complementary filter of green glass interposed in front of the camera lens. The exposure necessary is now increased greatly by the insensitiveness of the plate to green light, and had to be as much as 25 minutes. It will be noticed that the polarisation of the track as shown by the difference in brightness of the two polarised images is quite as prominent in Fig. 2 as in Fig. 1.

I may also mention that Mr. Krishnan and myself have succeeded in detecting the new radiation and observing its partial polarisation in a number of organic vapours and also in the gases $CO_2$ and $N_2O$. The problem in these cases is one of securing sufficient intensity of scattering for the effect to be detectable through the complementary filter. This can be secured by heating up the substance in a sealed bulb or by using steel observation-vessels for containing the compressed gases, so as to obtain sufficient density of the scattering molecules. The question of the background against which the track is observed is also of great importance.

The new type of secondary radiation is also observable in crystals such as ice, and in amorphous solids. It is thus a phenomenon whose universal nature has to be recognised.

### 4. *Line-Spectrum of New Radiation.*

That the secondary radiation passes the complementary filter and yet is strongly polarised to an extent comparable with the ordinary molecular scattering, is clear evidence that we have in it an entirely new type of secondary radiation which is distinct from either the ordinary scattering or the usual type of fluorescence. A striking and even startling confirmation of this view is furnished by an examination of its spectrum. Preliminary observations with sunlight filtered through a combination which passes a narrow range of wavelengths, showed the spectrum of the new radiation to consist mainly of a narrow range of wave-lengths clearly separated from the incident spectrum by a dark space. This encouraged me to take up observations with a monochromatic source of light. A quartz mercury lamp with a filter which completely cuts out all the visible lines of longer wave-length than the indigo line 4,358. A. U. was found to be very effective. When the light from such a lamp was passed through the bulb containing a dust-free liquid, and the spectrum of the scattered light was observed through a direct-vision spectroscope, it was found to exhibit two or more sharp bright lines in the blue and green regions of the spectrum. These lines are not present in the spectrum of the incident light or in the unfiltered light of the mercury arc and are thus manufactured by the molecules of the liquid.

Figs. 3 (1) and 3 (2), and Figs. 4 (1) and 4 (2) show the phenomenon. They are spectrograms taken with a small Hilger quartz instrument of the scattering by *liquid* benzene. Fig. 3 was taken with the light from the quartz mercury arc filtered through a blue glass which allows the wave-lengths from about 3,500 A. U. to 4,400 A. U. to pass through. Fig. 3 (1) represents the incident-spectrum and Fig. 3 (2) the scattered spectrum, and the latter shows a number of sharp lines not present in Fig. 3 (1). These are indicated in the figure. Figs. 4 (1) and (2) similarly represent the incident and scattered spectra with benzene liquid, the filter used being a potassium permanganate solution. Here again the new lines which appear are indicated in the figure. Visual observations were also made using a quinine sulphate solution together with the blue glass as a filter and thus cutting off all the radiations except 4,358 A.U. from the incident spectrum. Some of the modified lines then disappear, leaving only those of longer wave-length. It is thus clear that each line in the incident spectrum gives rise to at least two lines in the scattered spectrum, one in the original or unmodified position, and a second in a shifted position of longer wave-length. There is thus a striking analogy with the Compton effect in the X-ray region.

There has, as yet, not been sufficient time for photographing the spectra from a large number of liquids, or even for measuring the photographs already obtained. Visual observations have however been made with a large number of liquids There is an astonishing similarity between the spectra obtained with different liquids. When only the 4,358 line was used, most liquids showed in the spectrum of the scattered light, a bright line in the blue-green region of the spectrum (about 5,000 A.U.), whose position was practically the same for chemically similar liquids such as pentane, hexane and octane for instance. There was, however, a recognizable difference in the position of the modified line when other liquids such as benzene or water were used. When the 4,047 line of the mercury arc was let in by removing the quinine sulphate solution, a second modified line in the blue region of the spectrum was seen with most liquids.

Photographs obtained so far with benzene and toluene suggest that there may be several modified lines, and that each modified line may be a doublet in some cases. In many liquids, the scattered spectrum shows in addition to sharp lines also an unmistakable continuous spectrum accompanying it. Carbon disulphide behaves in an exceptional manner, showing a diffuse band.

Observations already made show that the new lines in the scattered spectrum are usually markedly polarised ; they also suggest that a continuous spectrum, when present, is less markedly polarised.

## 5. *Nature of the New Radiation.*

The discovery set out above naturally opens up an array of problems for investigation. The most pressing question is, how is the modified scattered radiation, as we may call it, generated by the molecules of the liquid ? As a tentative explanation, we may adopt the language of the quantum theory, and say that the incident quantum of radiation is partially absorbed by the molecule, and that the unabsorbed part is scattered. The suggestion does not seem to be altogether absurd and indeed such a possibility is already contemplated in the Kramers-Heisenberg theory of dispersion. If we accept the idea indicated above, then the difference between the incident and scattered quanta would correspond to a quantum of absorption by the molecule. The measurement of the frequencies of the new spectral lines thus opens a new pathway of research into molecular spectra, particularly those in the infra-red region.

If a molecule can take up part of the incident quantum of radiation and scatter the remaining part, then it might also be capable of adding a quantum of its own characteristic frequency to the incident radiation when scattering it. In such a case we should expect a modified line of *increased* frequency. Such a result appears to be shown in Fig. 3 (2) of Plate XII, as a solitary line in the extreme left of the photograph. This result, however, requires to be confirmed by more photographs and with other liquids. So far it would appear that a degradation of frequency is more probable than an enhancement. It is too early to speculate at present on the origin of the continuous radiation observed in some cases, whether it is due to changes in the molecule itself, or whether it arises from inelastic collisions of the second kind within the liquid resulting in *partial* transformation of the incident quantum of radiation into translatory kinetic energy of the molecules. When further data are obtained, it should be possible to express a definite opinion on this point, and also on the role played by the solvent in the explanation of ordinary fluorescence.

## 6. *Relation to Thermodynamics.*

As explained in the introduction, the ordinary scattering of light can be regarded equally well as a molecular effect, and as a bulk effect arising from the thermodynamic fluctuations of the whole medium. The question arises whether the new type of secondary radiation is exclusively a molecular effect or not, and whether it is related in any way to thermodynamics. The question is obviously one to be answered by experiment and theory conjointly. The comparative study of the effect at different temperatures and in different states of aggregation of matter is obviously of great importance in this connection. It has already been remarked that the effect is observable in gases and vapours and indeed it is found possible to determine its intensity and polarisation in the gaseous state. It is also of great interest to remark that the solid crystal ice also shows the sharp modified lines in the scattered spectrum in approximately the same positions as pure water. The only observations made with amorphous solids are with optical glass. Here the modified scattered spectrum consists of diffuse bands and not sharp lines. Whether this is generally true for all amorphous solids, and whether any changes occur at low and high temperatures remains to be determined by experiment.

### 7. *Coherent or Non-Coherent Radiation ?*

An important question to be decided in the first instance by experiment is whether the modified scattered radiations from the different molecules are incoherent with each other. One is tempted to assume that this must be the case, but a somewhat astonishing observation made with liquid carbon dioxide contained in steel observation vessels gives us pause here. It was found on blowing off the $CO_2$ by opening a stopcock, a cloud formed within the vessels which scattered light strongly in the ordinary way. On viewing the cloud through the complementary filter, the scattered radiation of modified frequency also brightened up greatly. This would suggest that the assumption of non-coherence is unjustifiable. Further, some qualitative observations suggest that the modified scattering by a mixture of carbon disulphide and methyl alcohol also brightens up notably at the critical solution temperature. Quantitative observations are necessary to decide the very fundamental question here raised.

## 8. *Possible X-Ray Analogies.*

If a quantum of radiation can be absorbed in part and scattered in part in the optical region of the spectrum, should not similar phenomena also occur in X-ray scattering? The type of scattering discovered by Prof. Compton may possibly be only one of numerous other types of scattering with modified frequencies, some with a line spectrum and some in the nature of continuous radiation. The extreme ultra-violet region of the spectrum may also furnish us with numerous examples of the new type of radiation, which clearly occupies a position intermediate between scattering and flourescence.

## 9. *Conclusion.*

We are obviously only at the fringe of a fascinating new region of experimental research which promises to throw light on diverse problems relating to radiation and wave-theory, X-ray optics, atomic and molecular spectra, flourescence and scattering, thermodynamics and chemistry. It all remains to be worked out.

I have to add in conclusion that I owe much to the valuable co-operation in this research of Mr. K. S. Krishnan, and the assistance of Mr. S. Venkateswaran and other workers in my laboratory.

The line spectrum of the new radiation was first seen on the 28th February, 1928. The observation was given publicity the following day.

*(Issued separately, 31st March, 1928).*

[P13]

# THE DIFFRACTION OF LIGHT BY HIGH FREQUENCY SOUND WAVES: PART I.

BY C. V. RAMAN
AND
N. S. NAGENDRA NATH.
*(From the Department of Physics, Indian Institute of Science, Bangalore.)*

Received September 28, 1935.

## 1. *Introduction.*

As is well known, Langevin showed that high frequency sound-waves of great intensity can be generated in fluids by the use of piezo-electric oscillators of quartz. Recently, Debye and Sears[1] in America and Lucas and Biquard[2] in France have described very beautiful experiments illustrating the diffraction of light by such high-frequency sound-waves in a liquid. Amongst the experimenters in this new field, may be specially mentioned R. Bär[3] of Zürich who has carried out a thorough investigation and has published some beautiful photographs of the effect. The arrangement may be described briefly as follows. A plane beam of monochromatic light emerging from a distant slit and a collimating lens is incident normally on a cell of rectangular cross-section and after passing through the medium emerges from the oppsite side. Under these conditions, the incident beam will be undeviated if the medium be homogeneous and isotropic. If, however, the medium be traversed by high-frequency sound-waves generated by introducing a quartz oscillator at the top of the cell, the medium becomes stratified into parallel layers of varying refractive index. Considering the case in which the incident beam is parallel to the plane of the sound-waves, the emerging light from the medium will now consist of various beams travelling in different directions. If the inclination of a beam with the incident light be denoted by $\theta$, it has been found experimentally that the formula

$$\sin\theta = \pm\frac{n\lambda}{\lambda^*}, \quad n \text{ (an integer)} \geqslant 0 \tag{1}$$

is in satisfactory agreement with the observed results, where $\lambda$ and $\lambda^*$ are the wave-lengths of the incident light and the sound wave in the medium

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[1] P. Debye and F. W. Sears, *Proc. Nat. Acad. Sci.* (Washington), 1932, **18**, 409.

[2] R. Lucas and P. Biquard, *Jour. de Phys. et Rad.*, 1932, **3**, 464.

[3] R. Bär, *Helv. Phys. Acta*, 1933, **6**, 570.

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respectively. With sound waves of sufficient intensity, numerous orders of these diffraction spectra have been obtained ; a wandering of the intensity amongst these orders has also been noticed by Bär[3] when the experimental conditions are varied.

Various theories of the phenomena have been put forward by Debye and Sears,[1] by Brillouin,[4] and by Lucas and Biquard.[2] The former have not presented quantitative results and it is hard to understand from their theory as to why there should be so many orders and why the intensity should wander between the various orders under varying experimental conditions. In Brillouin's theory, the phenomenon is attributed to the reflection of light from striations of the medium caused by the sound waves. We know, however, from the work of Rayleigh that the reflection of light by a medium of varying refractive index is negligible if the variation is gradual compared with the wave-length of light. Under extreme conditions, we might perhaps obtain the Brillouin phenomenon, but the components of reflection should be very weak in intensity compared to the transmitted ones. As one can see later on in this paper, the whole phenomenon including the positions of the diffracted beams and their intensities can be explained by a simple consideration of the transmission of the light beam in the medium. Lucas and Biquard attribute the phenomenon to an effect of mirage of light waves in the medium. In what way the relation (1) enters in their theory is not clear. The wandering of the intensities of the various components observed by Bär has not found explanation in any of the above theories.

We propose in this paper a theory of the phenomenon on the simple consideration of the regular transmission of light in the medium and the phase changes accompanying it. The treatment is limited to the case of normal incidence. The formula (1) has been established in our theory. Also, a formula for the intensities of the various components has been derived. It is found that the above results are in conformity with the experimental results of Bär.[3]

## 2. *Diffraction of light from a corrugated wave-front.*

The following theory bears a very close analogy to the theory of the diffraction of a plane wave (optical or acoustical) incident normally on a periodically corrugated surface, developed by the late Lord Rayleigh.[5] He showed therein that a diffraction phenomenon would ensue in which the positions of the various components are given by a formula similar to (1)

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[4] L. Brillouin, "La Diffraction de la Lumiere par des Ultra-sons", *Act. Sci. et Ind.*, 1933, 59.

[5] Lord Rayleigh, *Theory of Sound* (Vol. 2), page 89.

and their relative intensities are given by a formula similar to the one we have found.

Consider a beam of light with a plane wave-front emerging from a rectangular slit and falling normally on a plane face of a medium with a rectangular cross-section and emerging from the opposite face parallel to the former. If the medium has the same refractive index at all its points, the incident beam will emerge from the opposite face with its direction unchanged. Suppose we now create layers of varying refractive index in the medium, say by suitably placing a quartz oscillator in the fluid. If the distance between the two faces be small, the incident light could be regarded as arriving at the opposite face with variations in the phase at its different parts corresponding to the refractive index at different parts of the medium. The change in the phase of the emerging light at any of its parts could be simply calculated from the optical lengths found by multiplying the distance between the faces and the refractive index of the medium in that region. This step is justified for $\int \mu(x, y, z)ds$ taken over the actual path is minimum, *i.e.*, it differs from the one taken over a slightly varied hypothetical path by a differential of the second order. So, the incident wave-front becomes a periodic corrugated wave-front when it traverses a medium which has a periodic variation in its refractive index. The origin of the axes of reference is chosen at the centre of the incident beam projected on the emerging face, the boundaries of the incident beam being assumed to be parallel to the boundaries of the face. The X-axis is perpendicular to the sound-waves and the Z-axis is along the direction of the incident beam of light. If the incident wave is given by

$$Ae^{2\pi i\nu t}$$

it will be

$$Ae^{2\pi i\nu\{t - L\mu(x)/c\}}$$

when it arrives at the other face where L is the distance between the two faces and $\mu(x)$ the refractive index of the medium at a height $x$ from the origin. It is assumed that the radii of curvature of the corrugated wave-front are large compared with the distance between the two faces of the cell. If $\mu_0$ be the refractive index of the whole medium in its undisturbed state, we can write $\mu(x)$ as given by the equation

$$\mu(x) = \mu_0 - \mu \sin \frac{2\pi x}{\lambda^*}$$

ignoring its time variation, $\mu$ being the ***maximum variation*** of the refractive index from $\mu_0$.

The amplitude due to the corrugated wave at a point on a distant screen parallel to the face of the medium from which light is emerging whose join with the origin has its $x$-direction-cosine $l$, depends on the evaluation of the diffraction integral

$$\int_{-p/2}^{p/2} e^{2\pi i\{lx+\mu L \sin(2\pi x/\lambda^*)\}/\lambda}\,dx$$

where $p$ is the length of the beam along the X-axis. The real and the imaginary parts of the integral are

$$\int_{-p/2}^{p/2}\{\cos ulx \cos(v \sin bx) - \sin ulx \sin(v \sin bx)\}dx$$

and

$$\int_{-p/2}^{p/2}\{\sin ulx \cos(v \sin bx) + \cos ulx \sin(v \sin bx)\}dx$$

where $u = 2\pi/\lambda$, $b = 2\pi/\lambda^*$ and $v = u\mu L = 2\pi\mu L/\lambda$.

We need the well-known expansions

$$\cos(v \sin bx) = 2\sum_0^\infty{}' J_{2r} \cos 2rbx$$

$$\sin(v \sin bx) = 2\sum_0^\infty J_{2r+1} \sin \overline{2r+1}\,bx$$

to evaluate the integrals, where $J_n[= J_n(v)]$ is the Bessel function of the $n$th order and a dash over the summation sign indicates that the coefficient of $J_0$ is half that of the others. The real part of the integral is then

$$2\sum_0^\infty{}' J_{2r}\int_{-p/2}^{p/2}\cos ulx \cos 2rbx\,dx - 2\sum_0^\infty J_{2r+1}\int_{-p/2}^{p/2}\sin ulx \sin \overline{2r+1}\,bx\,dx$$

or

$$\sum_0^\infty{}' J_{2r}\int_{-p/2}^{p/2}\{\cos(ul+2rb)x + \cos(ul-2rb)x\}dx$$

$$+\sum_0^\infty J_{2r+1}\int_{-p/2}^{p/2}\{\cos(ul+\overline{2r+1}\,b)x - \cos(ul-\overline{2r+1}\,b)x\}dx$$

Integrating the above, we obtain

$$p\sum_0^\infty{}' J_{2r}\left\{\frac{\sin(ul+2rb)p/2}{(ul+2rb)p/2} + \frac{\sin(ul-2rb)p/2}{(ul-2rb)p/2}\right\}$$

$$+p\sum_0^\infty J_{2r+1}\left\{\frac{\sin(ul+\overline{2r+1}\,b)p/2}{(ul+\overline{2r+1}\,b)p/2} - \frac{\sin(ul-\overline{2r+1}\,b)p/2}{(ul-\overline{2r+1}\,b)p/2}\right\} \quad \cdot\cdot \quad (2)$$

The integral corresponding to the imaginary part of the diffraction integral is zero. One can see that the magnitude of each individual term of (2) attains its highest maximum (the other maxima being negligibly small compared to the highest) when its denominator vanishes. Also, it can be seen that when any one of the terms is maximum, all the others have negligible values as the numerator of each cannot exceed unity and the denominator is some integral non-vanishing multiple of $b$ which is sufficiently large. So the maxima of the magnitude of (2) correspond to the maxima of the magnitudes of the individual terms. Hence the maxima occur when

$$ul \pm nb = 0 \quad n(\text{an integer}) \geqslant 0 \quad \dots \quad \dots \quad \dots \quad \dots \quad (3)$$

where $n$ is any even or odd positive integer. The equation (3) gives the directions in which the magnitude of the amplitude is maximum which correspond also to the maximum of the intensity. If $\theta$ denotes the angle between such a direction in the XZ-plane along which the intensity is maximum and the direction of the incident light, (3) can be written as

$$\sin \theta = \pm \frac{n\lambda}{\lambda^*} \quad \dots \quad \dots \quad \dots \quad \dots \quad \dots \quad (4)$$

remembering that $u = 2\pi/\lambda$ and $b = 2\pi/\lambda^*$. This formula is identical with the formula (1) given in the first section. The magnitudes of the various components in the directions given by (4) can be calculated if we know,

$$J_n \text{ or } J_n(v) \text{ or } J_n(2\pi\mu L/\lambda).$$

Thus the relative intensity of the $m$th component to the $n$th component is given by

$$\frac{J_m^2(v)}{J_n^2(v)} \quad \text{where } v = 2\pi\mu L/\lambda.$$

In the undisturbed state of the medium there is no variation of the refractive index, *i.e.*, $\mu = 0$. In this case all the components vanish except the zero component for

$$J_m(0) = 0 \text{ for all } m \neq 0 \text{ and } J_0(0) = 1.$$

In the disturbed state, the relative intensities depend on the quantity $v$ or $2\pi\mu L/\lambda$ where $\lambda$ is the wave-length of the incident light, $\mu$ is the maximum variation of the refractive index and L is the path traversed by light in the medium. We have calculated the relative intensities of the various components which are observable for values of $v$ lying between 0 and 8 at different steps (Fig. 1).

Fig. 1 shows that the number of observable components increases as the value of $v$ increases. When $v = 0$, we have only the central component. As $v$ increases from 0, the first orders begin to appear. As $v$ increases still more, the intensity of the central component decreases steadily and the first orders increase steadily in their intensity till they attain maximum intensity when the zero order will nearly vanish and the second orders will have just appeared. As $v$ increases still more, the zero order is reborn and increases

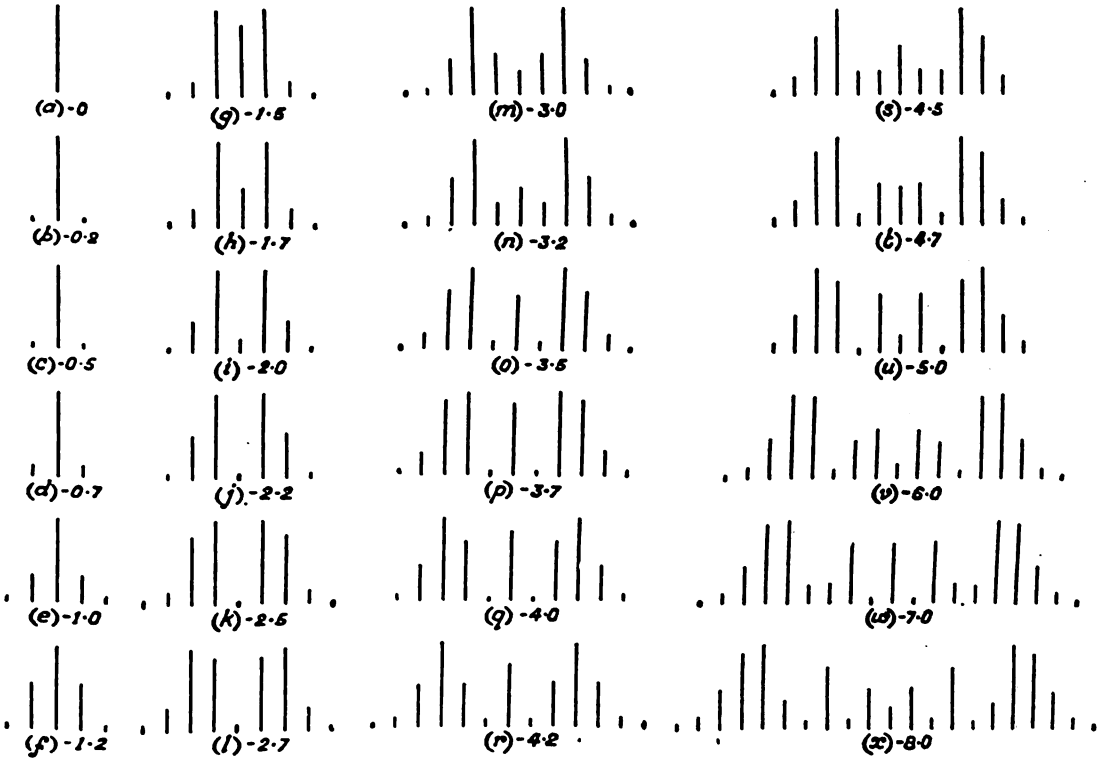

FIG. 1.

Relative intensities of the various components in the diffraction spectra.
(For tables, see Watson's *Bessel Functions and Report of the British Association*, 1915.)

in its intensity, the first orders fall in their intensity giving up their former exalted places to the second orders; while the third orders will have just appeared and so on.

Our theory shows that the intensity relations of the various components depend on the quantity $v$ or $2\pi\mu L/\lambda$. Thus an *increase* of $\mu$ (*i.e.*, an increase of the supersonic intensity which creates a greater variation in the refractive index of the medium) or an *increase* of L or a *decrease* of $\lambda$ should give similar effects *except* in the last case where the directions of the various beams will be altered in accordance with (4).

### 3. *Interpretation of Bär's Experimental Results.*

(*a*) *Dependence of the effect on the supersonic intensity.*—Bär has observed that only the zero order (strong) and the first orders (faint) are present when the supersonic intensity is not too great. He found that more orders appear as the supersonic intensity is increased but that the intensity of the zero order decreases while the first orders gain in their intensity. Increasing the supersonic intensity more, he found that the first order would become very faint while the second and third orders will have about the same intensity. The figures 1*a* of his paper may very well be compared with our figures 1(*c*), 1(*h*) and 1(*k*). Thus, we are able to explain the appearance of more and more components and the wandering of the intensity amongst them as the supersonic intensity is increased, in a satisfactory manner.

(*b*) *Dependence of the effect on the wave-length of the incident light.*—We have already pointed out that the effects due to an increase of $\mu$ caused by an increase of supersonic intensity are similar due to those with a decrease of $\lambda$ except for the fact that the positions of the components of the emerging light alter in accordance with (4). Bär has obtained two patterns of the phenomenon by using light with wave-lengths 4750Å and 3650Å. He obtained, using the former seven components and using the latter eleven components in all. He also observed great variations in the intensities of the components. Not only is the increase in the number of components an immediate consequence of our theory, but we can also find the pattern with 3650Å if we assume the pattern with 4750Å. The pattern with the latter in Bär's paper shows a strong resemblance to our figure 1(*p*) for which $2\pi\mu L/\lambda$ is 3·7. Thus we can calculate $2\pi\mu L/\lambda$ when $\lambda$ is 3650Å. It comes to about 4·8. Actually our figure for which $2\pi\mu L/\lambda$ is 4·8 closely corresponds to Bär's pattern with 3650Å.

(*c*) *Dependence of the effect on the length of the medium which the light traverses.*—It is clear from our theory that an increase of L corresponds to an increase of $v$ and that the effects due to this variation would be similar to those with an increase of the supersonic intensity. But the basis of our theory does not actually cover any large change in L. However, we should find more components and the wandering of the intensity amongst the various components.

## 4. *Summary.*

(*a*) A theory of the phenomenon of the diffraction of light by sound-waves of high frequency in a medium, discovered by Debye and Sears and Lucas and Biquard, is developed.

(*b*) The formula

$$\sin\theta = \pm \frac{n\lambda}{\lambda^*} \quad n \text{ (an integer)} \gg 0$$

which gives the directions of the diffracted beams from the direction of the incident beam and where $\lambda$ and $\lambda^*$ are the wave-lengths of the incident light and the sound wave in the medium, is established. It has been found that the relative intensity of the $m$th component to the $n$th component is given by

$$J_m^2(2\pi\mu L/\lambda) \;/\; J_n^2(2\pi\mu L/\lambda)$$

where the functions are the Bessel functions of the $m$th order and the $n$th order, $\mu$ is the maximum variation of the refractive index and L is the path traversed by light. These theoretical results interpret the experimental results of Bär in a very gratifying manner.

# THE DIFFRACTION OF LIGHT BY SOUND WAVES OF HIGH FREQUENCY: PART II.

By C. V. Raman

and

N. S. Nagendra Nath.

(*From the Department of Physics, Indian Institute of Science, Bangalore.*)

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## 1. *Introduction.*

In the first[1] of this series of papers, we were concerned with the explanation of the diffraction effects observed when a beam of light traverses a medium filled by sound waves of high frequency. For simplicity, we confined our attention to the case in which a plane beam of light is normally incident on a cell of the medium with rectangular cross-section and travels in a direction strictly perpendicular to the direction along which the sound waves are propagated in the medium. By taking into account the corrugated form of the wave-front on emergence from the cell, the resulting diffraction-effects were evaluated. This treatment will be extended in the present paper to the case in which the light waves travel in a direction inclined at a definite angle to the direction of the propagation of the sound waves. The extension is simple, but it succeeds in a remarkable way in explaining the very striking observations of Debye and Sears[2] who found a characteristic variation of the intensity of the higher orders of the diffraction spectrum when the angle between the incident beam of light and the plane of the sound waves was gradually altered.

We shall first set out a simple geometrical argument by which the changes in the diffraction phenomenon which occur with increasing obliquity can be inferred from the results already given for the case of the normal incidence. An analytical treatment then follows which confirms the results obtained geometrically.

## 2. *Elementary Geometrical Treatment.*

The following diagrams illustrate the manner in which the amplitude of the corrugation in the emerging wave-front alters as the incidence of light on the planes of the sound waves is gradually changed. In the diagrams,

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[1] C. V. Raman and N. S. Nagendra Nath, *Proc. Ind. Acad. Sci.*, 1935, 2, 406–412.

[2] P. Debye and F. W. Sears, *Proc. Nat. Acad. Sci.* (Washington), 1932, 18, 409.

the planes of maximum and minimum density caused by the sound waves at any instant of time are indicated by thick and thin lines (*e.g.*, AB and CD) respectively. The paths of the light rays are represented by dotted lines in Figs. 1 (*b*), (*c*) and (*d*). As we are mainly interested in the calculation of the phase-changes which the incident wave undergoes before it emerges from the cell, the bending of the light rays within the medium may, in virtue of Fermat's well-known principle, be ignored without a sensible error, *provided* the total depth of the cell is not excessive.

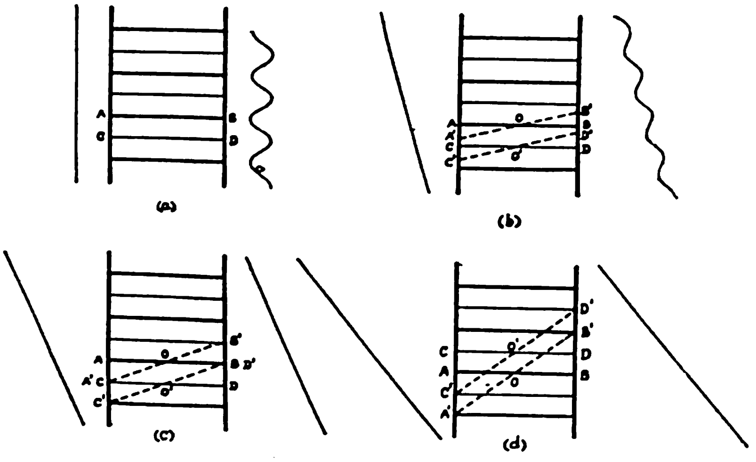

FIG. 1.

Considering the variation in the refractive index to be simply periodic, the neighbouring light-paths with maximum and minimum optical lengths AB and CD respectively, in the case of normal incidence, are shown in Fig. 1(*a*). The lines AB and CD are separated by $\lambda^*/2$ where $\lambda^*$ is the wave-length of the sound waves. The difference between the maximum and the minimum optical lengths gives a measure of the corrugation of the wave-front on emergence. Considering now a case in which the light rays make an angle $\phi$ with the planes of the sound waves, we may denote the maximum and the minimum optical lengths by A'B' and C'D' respectively. These would be symmetrically situated with respect to AB and CD, and would tend to coincide with them as $\phi$ is decreased. The optical length of A'B' is *less* than that of AB, for the refractive index at any point except at O is less than the constant maximum refractive index along AB, $\phi$ being small. On the other hand, the optical length of C'D' is *greater* than that of CD, for the refractive index is minimum along CD. A simple consideration of the above shows that the difference between the optical lengths of A'B' and C'D' is less than that between those of AB and CD. As this difference gives twice the amplitude of the corrugation of the emerging wave-front, it follows, in the case shown in Fig. 1 (*b*), that the amplitude of the corrugation of the emerging wave-front is less than that in the case of Fig. 1 (*a*).

Fig. 1 (*c*) illustrates a case when the maximum optical length is just equal to the minimum optical length. This occurs when the direction of the incident beam is inclined to the planes of the sound-wave-fronts at an angle $\alpha_1$ given by $\tan^{-1}\frac{\mathrm{B'B}}{\mathrm{OB}} = \tan^{-1}\frac{\lambda^*/2}{\mathrm{L}/2} = \tan^{-1}(\lambda^*/\mathrm{L})$. That the optical lengths of A′B′ and C′D′ in Fig. 1 (*c*) are equal follows by a very simple geometrical consideration. Thus, when light rays are incident on the sound waves at an angle $\tan^{-1}(\lambda^*/\mathrm{L})$, the amplitude of the corrugation of the emerging wave-front vanishes, *i.e.*, a plane incident beam of light remains so when it emerges from the medium. This result would also be true whenever $\alpha_n = \tan^{-1}(n\lambda^*/\mathrm{L})$, $n \neq 0$. The case when $n=2$ is illustrated in Fig. 1 (*d*). In all these cases the diffraction effects disappear. As the corrugation vanishes when $\phi$ is $\alpha_{n+1}$ or $\alpha_n$, there is an intermediate direction which makes an angle $\beta_n$ with the sound waves giving the maximum corrugation if light travels along that direction. We can take $\beta_0(=0)$ to represent the case when the incident beam of light is parallel to the sound waves.

Thus, we have deduced that the corrugation of the emerging wave-front is maximum when the direction of light is parallel to the sound waves $[\beta_0(=0)]$, decreases steadily to zero as the inclination $\phi$ between the incident light and the sound waves is increased to $\alpha_1$, increases to a smaller maximum as $\phi$ increases from $\alpha_1$ to $\beta_1$, decreases to zero as $\phi$ increases from $\beta_1$ to $\alpha_2$, increases to a still smaller maximum as $\phi$ increases from $\alpha_2$ to $\beta_2$, and so on. (I)

As the variation of the refractive index is simply periodic along the direction normal to the sound-wave-fronts, it follows that the optical length of the light path is also simply periodic along the same direction when the incident light rays are parallel to the sound waves. This means that the corrugation of the emerging wave-front is also simply periodic. When the incident light rays are incident at an angle $\phi$ to the sound waves, the optical length of the light path would be simply periodic in a direction perpendicular to the light rays. This means that the emerging wave-front would be tilted by the angle $\phi$ about the line of the propagation of the sound waves and that its corrugation would be simply periodic along the same line.

We have shown in our previous paper that a simply periodic corrugated wave is equivalent to a number of waves travelling in directions which make angles, denoted by $\theta$, with the direction of the incident beam given by

$$\sin\theta = \pm \frac{n\lambda}{\lambda^*} \qquad n \text{ (an integer)} > 0 \qquad .. \quad (1)$$

where $\lambda$ is the wave-length of the incident light. In view of the results obtained in the previous paragraph, the formula (1) would also hold good when the incident light is a small angle with the sound waves.

The relative intensities of the various diffraction spectra which depend on the amplitude of the corrugation should obey a law similar to the one in the case of the normal incidence.

Thus, we find that the results in the case of an oblique incidence would be similar to those of the normal incidence with the amplitude of the corrugation modified. Hence, we deduce, in virtue of the statement I, the following results, assuming the results, in the case of normal incidence, obtained in our earlier paper.

The diffraction spectrum will be most prominent when $\phi = 0$. The intensity of the various components wander when $\phi$ is increased. When $\phi$ increases from zero to $\alpha_1$, the number of the observable orders in practice decreases and when $\phi = \alpha_1$ all the components disappear except the central one which will attain maximum intensity. This does not mean that the intensities of all the orders except the central one decrease to zero monotonically as $\phi$ varies from zero to $\alpha_1$, but some of them may attain maxima and minima in their intensities before they attain the zero intensity when $\phi = \alpha_1$. This is obvious in virtue of the property that the intensity of the $n$th component depends on the square of the Bessel function $J_n$. As $\phi$ increases from $\alpha_1$ to $\beta_1$ the intensity of the central component falls and the other orders are reborn one by one. As $\phi$ increases from $\beta_1$ to $\alpha_2$, the number of observable orders decreases and when $\phi = \alpha_2$ all the orders vanish except the central one which will attain the maximum intensity and so on.

### 3. *Analytical Treatment.*

In the following, we employ the same notation as in our earlier paper. The optical length of a path in the medium parallel to the direction of the incident light making an angle $\phi$ with the sound waves may be easily calculated. It is

$$\int_0^{L\sec\phi} \mu(s)ds$$

or

$$\mu_0 L \sec\phi - \mu \int_0^{L\sec\phi} \sin b(x - s\sin\phi)ds.$$

Integrating we obtain the integral as

$$\mu_0 L \sec\phi - \frac{\mu}{b\sin\phi}\{\sin(bL\tan\phi)\sin bx + [\cos(bL\tan\phi)-1]\cos bx\}.$$

The last term can be written as

$$-A\sin bx + B\cos bx$$

where

$$A = \frac{\mu}{b\sin\phi}\sin(bL\tan\phi)$$

$$B = -\frac{\mu}{b\sin\phi}[\cos(bL\tan\phi)-1].$$

Thus the optical length of the path can be written as

$$\mu_0 L \sec\phi - \sqrt{(A^2+B^2)}\sin b\left(x - \tan^{-1}\frac{B}{A}\right).$$

Ignoring the constant phase factor, the optical length is

$$\mu_0 L \sec\phi - \frac{2\mu}{b\sin\phi}\sin\left(\frac{bL\tan\phi}{2}\right)\sin bx.$$

If the incident light is

$$\exp\left[2\pi i\nu\left(t - \frac{x\sin\phi}{c}\right)\right]$$

when it arrives at the face of the cell, it will be

$$\exp\left[\frac{2\pi i}{\lambda}\left(ct - x\sin\phi - \int_0^{L\sec\phi}\mu(s)ds\right)\right]$$

when it arrives at the face from which it emerges.

The amplitude of the corrugated wave at a point on the screen whose join with the origin has its $x$-direction-cosine $l$, depends on the evaluation of the diffraction integral

$$\int_{-p/2}^{p/2}\exp\left[\frac{2\pi i}{\lambda}\left\{(l-\sin\phi)x + \frac{2\mu}{b\sin\phi}\sin\left(\frac{bL\tan\phi}{2}\right)\sin bx\right\}\right]dx.$$

The evaluation of the integral and the discussion of its behaviour with respect to $l$ may be effected in the same way as in our earlier paper. Maxima of the intensity due to the corrugated wave occur in directions making angles, denoted by $\theta$, with the direction of the incident beam when

$$\sin(\theta+\phi) - \sin\phi = \pm\frac{n\lambda}{\lambda^*} \qquad n \text{ (an integer)} > 0 \qquad \text{.. (1)}$$

The relative intensity of the $m$th order to the $n$th order is given by

$$\frac{J_m^2(v)}{J_n^2(v)} \qquad \text{.. .. .. .. .. (2)}$$

where

$$v = \frac{2\pi}{\lambda} \cdot \frac{2\mu}{b \sin \phi} \sin\left(\frac{b\mathrm{L} \tan \phi}{2}\right)$$

$$= \frac{2\pi\mu\mathrm{L}}{\lambda} \sec \phi \, \frac{\sin t}{t} \text{ where } t = \frac{b\mathrm{L} \tan \phi}{2} = \frac{\pi\mathrm{L} \tan \phi}{\lambda^*}.$$

The expression for the relative intensities in our earlier paper can be obtained from (2) by making $\phi \to 0$ when $v \to \frac{2\pi\mu\mathrm{L}}{\lambda} = v_0$. So the expression for the relative intensities

$$\mathrm{J}_m^2(v_0)/\mathrm{J}_n^2(v_0) \quad .. \quad .. \quad .. \quad .. \quad (3)$$

in the case of normal incidence will change to

$$\mathrm{J}_m^2(v)/\mathrm{J}_n^2(v)$$

where

$$v = v_0 \sec \phi \, \frac{\sin t}{t} \quad .. \quad .. \quad .. \quad .. \quad (4)$$

and

$$t = \frac{\pi\mathrm{L} \tan \phi}{\lambda^*}.$$

Even if $\phi$ be small so that $\sin \phi \simeq \tan \phi \simeq \phi$, it is *not* justifiable to write $\sin t \simeq t$ *unless* $\pi\mathrm{L}\phi/\lambda^*$ is also small to admit the approximation. As $\pi\mathrm{L}/\lambda^*$ is sufficiently large we should expect great changes in the diffraction phenomenon even if $\phi$ be a fraction of a degree. $v$ vanishes when

$$t = n\pi \qquad n \text{ (an integer)} > 0,$$

that is, when $\mathrm{L} \tan \phi = n\lambda^*$,

or

$$\phi = \tan^{-1} \frac{n\lambda^*}{\mathrm{L}}, \; n \text{ (an integer)} > 0,$$

confirming the same result obtained geometrically. Whenever $v$ vanishes, it can be seen that the amplitude of the corrugation of the wave-front also vanishes. The statement I in Section 2 and the consequences with regard to the behaviour of the intensity among the various orders can all be confirmed by the expression (3).

In the numerical case when L = 1 cm., and $\lambda^* = 0{\cdot}01$ cm., the amplitude of the corrugation vanishes $\tan \alpha_1 = 0{\cdot}01$ or $\alpha_1 = 0° \, 34'$. This means that as $\phi$ varies from 0° to 0° 34′, the relative intensities of the various orders wander according to (2) till when $\phi = 0° \, 34'$, all the orders disappear except the central one which attains maximum intensity. This does not mean that the intensities of all the orders except the central one decrease monotonically to zero but they *may possess* several maxima and minima before they become zero. The intensity of the $n$th order depends on the behaviour of $J_n^2\left[v_0 \sec\phi \,\frac{\sin(\pi L \tan\phi/\lambda^*)}{(\pi L \tan\phi/\lambda^*)}\right]$ under the above numerical conditions as $\phi$ varies from 0° to 0° 34′. As $\phi$ just exceeds 0° 34′, all the orders are reborn one by one till a definite value of $\phi$ after which they again fall one by one and when $\phi = 1°\, 8'$, all the orders disappear except the central one.

The numerical example in the above paragraph shows the delicacy of the diffraction phenomenon. If the wave-length is quite small, the diffraction phenomenon will be present in the case of the strictly normal *incidence* as the relative intensity expression (3) does not depend on $\lambda^*$ but will soon considerably change even for slight variations of $\phi$ as the relative intensity expression (4) depends on $\lambda^*$. One should be very careful in carrying out the intensity measurements in the case of normal incidence, for even an error of a few minutes of arc in the incidence will affect the intensities of the various orders.

### 4. *Comparison with the experimental results of Debye and Sears.*

Debye and Sears make the following statement in their paper: "Fixing the attention on one of the spectra *preferably of higher order*, one can observe that it attains its maximum intensity if the trough is turned through a small angle such that the primary rays are no longer parallel to the planes of the supersonic waves. Different settings are required to obtain highest intensities in different orders. If the trough is turned continuously in one direction, starting from a position which gave the highest intensity to one of the orders, the intensity decreases steadily, goes through zero, increases to a value much smaller than the first maximum, decreases to zero a second time and goes up and down again through a still smaller maximum." This statement very aptly describes the behaviour of the function

$$J_n^2\left[v_0 \sec\phi \,\frac{\sin(\pi L \tan\phi/\lambda^*)}{(\pi L \tan\phi/\lambda^*)}\right]$$

as $\phi$ alters under the conditions imposed in the above statement. The zeroes and the maxima of the intensity of the $n$th order, as a function of $\phi$, correspond to the zeroes and the maxima of the above function.

### 5. *Summary.*

The theory of the diffraction of light by sound waves of high frequency developed in our earlier paper is extended to the case when the light beam is incident at an angle to the sound wave-fronts, both from a geometrical point of view and an analytical one. It is found that the maxima of intensity of the diffracted light occur in directions which make definite angles, denoted by $\theta$, with the direction of the incident light given by

$$\sin(\theta+\phi) - \sin\phi = \pm \frac{n\lambda}{\lambda^*},\ n \text{ (an integer)} \geqslant 0$$

where $\lambda$ and $\lambda^*$ are the wave-lengths of the incident light and the sound waves in the medium. The relative intensity of the $m$th order to the $n$th order is given by

$$J_m^2\left(v_0 \sec\phi \frac{\sin t}{t}\right) \Big/ J_n^2\left(v_0 \sec\phi \frac{\sin t}{t}\right)$$

where $v_0 = \dfrac{2\pi\mu L}{\lambda}$, $t = \dfrac{\pi L \tan\phi}{\lambda^*}$, $\phi$ is the inclination of the incident beam of light to the sound waves, $\mu$ is the maximum variation of the refractive index in the medium when the sound waves are present and $L \sec\phi$ is the distance of the light path in the medium. These results explain the variations of the intensity among the various orders noticed by Debye and Sears for variations of $\phi$ in a very gratifying manner.

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