RESEARCH WITH STYLE: THE STORY OF RAMAN'S STUDY OF LIGHT SCATTERING+

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ABSTRACT

Nature had a very profound impact on C.V. Raman and its contemplation influenced his scientific thinking. In early 1922, Raman, perceived that Rayleigh Scattering takes place in a discontinuous manner and that the quantum nature of light must play an important role in the phenomenon of light scattering. This led to his suggesting that Maxwell's field equations will have to be quantized. These ideas launched him on a series of experimental researches which culminated in the discovery of the Raman Effect. An attempt is made here to describe the atmosphere that existed in his laboratory during this period, the excitement Raman felt, the enthusiasm he transmitted to his students, and the thrill they experienced in working with him.

INTRODUCTION

To those of us who knew C. V. Raman well, what struck us most was his intense love of and preoccupation with Nature. In one of his lectures he said[1]:

> "The face of Nature as presented to us is infinitely varied; but to those who love her, it is ever beautiful and interesting. The blue of the sky, the glories of sunrise and sunset, the ever shifting panorama of the clouds, the varied colours of the forest and field and the star sprinkled sky at night — these and many other scenes pass before our eyes on the never ending drama of light and colour which Nature presents for our benefit".

In another lecture[1]:

> "The man of science observes what Nature offers with the eye of understanding but her beauties are not lost on him for that reason. More truly it can be said that understanding refines our vision and heightens our appreciation of what is striking and beautiful".

Clearly a different view from Goethe's who wrote that Newton's analysis of the rainbow colours "would cripple Nature's art". Among the natural phenomena that most fascinated Raman were the beautiful coronae and haloes one can see around the sun and the moon when thin clouds come in front of them. This fascination never ceased. In 1910, when he was an Assistant Accountant General in Nagpur, his clerks noticed him at lunch-time studying the solar coronae reflected in a pool of water in front of his office. Later in Calcutta, he was seen often making observations on the lunar coronae, when taking his evening stroll in the *maidan*. I myself have seen him measuring the polarization of the coronae in 1967 in Bangalore when he was 79! Wordsworth's lines on ***The Rainbow*** come to mind

So was it when my life began
So is it now that I am a man
So will it be when I shall grow old ......

Raman stated[2] that purely from the size of the rings, the vividness of the colours, and the polarization characteristics, one cannot only estimate the size and distribution of the parti-

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cles but also deduce whether these are droplets of liquid water, amorphous solidified water or ice crystals. Raman produced these coronae in the laboratory over a wide range of droplet sizes and states that these artificial coronae are more striking in colour than those seen in Nature, as the colours of the latter were diluted by the finite angular dimensions of the sun or moon[2]. He was often up very early in the morning at Bangalore observing the coronae formed around the planet Venus!

It is also remarkable how much science Raman could extract from his study and contemplation of this one phenomenon. I shall give three examples.

RAMAN'S OBSERVATION OF "SPECKLES" IN 1919*[2]

Who has not seen the *radiant spectra*—the rays that seem to emanate — when a small intense source of light is viewed against a dark background — the long coloured streamers of light which are seen to diverge from the source in all directions. Raman commenced his detailed studies of this phenomenon around 1918[3]. He noted that when the source is monochromatic, the streamers become spots and faint haloes encircle the source near the outer limit. He connected this phenomenon with the coronae.

Coronae round the sun and the moon are the result of Fraunhofer diffraction by spherical droplets. Raman argued that when radiations diffracted by the individual particles are superposed at any given point of observation, there must be interference. If the particles are distributed at random and execute rapid uncorrelated movements as in a real cloud, these interference effects will be unobservable. The observed intensity would be n times the Fraunhofer diffraction of an individual droplet (assuming n particles of equal size). If on the other hand, the particles occupy stationary positions within the cloud, the interferences between the individual particles must be observable, even if the particles are numerous. For a random distribution of particles, while the most probable resultant intensity would be zero, the average intensity would be nA^2 (where A is the amplitude of the wave scattered by one particle). Hence a point source of monochromatic light, when viewed through a stationary cloud, will exhibit a diffraction corona on which will be "superposed" this interference effect. The net result is that instead of a continuous distribution of intensity, one would observe violent fluctuations. The corona, to use Raman's expression must have a "mottled" appearance, each point or "mottle" being like an optical image of the original source produced by the cloud of particles! If the source is one of white light, each point is spread radially into a spectrum (the red being the farthest from the centre) giving the effect of long coloured streamers. The intensity distribution of the spots will be that obtained from the random walk problem i.e. the Rayleigh law.

Much later, Raman and his (now renowned) student G. N. Ramachandran verified all these deductions in coronae produced by a cloud of lycopodium particles on a glass plate. Incidentally the *mottles* of Raman are the 'speckles' which became prominent when the laser was discovered a few decades later. Raman used to project the halo produced by the colloidal particles of dilute milk and showed that the bright spots (speckles) in the central disk of the corona continually changed, appearing and disappearing in the field of view — an effect due to the Brownian motion of the colloidal particle. In 1919 Raman proposed that the diffracting structures which produced the *radiant spectra* were in the eye. They were in the refractive media of the eye-opaque or transparent particles or small regions with small differences in refractive index in the cornea, the vitreous or aqueous humour or even the lens. These ideas which he put forth, anticipated, in a sense, the modern concepts of

* A consolidated account of these researches were given in the "Sayaji Rao Gaekwad Foundation Lectures" by Raman in February 1941. Unfortunately only a part of it was published as "Lectures on Physical Optics"[2] in 1959. This is reproduced in the "Scientific Papers of C. V. Raman—Vol. III Optics", Indian Academy of Sciences (in press).

twinkling of stars and the speckle formation in telescopes — a connection which was even hinted at by him.

X-RAY DIFFRACTION BY LIQUIDS

It is almost certain that Raman's interest in crystal structure determination and in X-ray diffraction arose due to his meeting Sir William Bragg (Senior) in London. Bragg showed him the structure of naphthalene that he had determined. Raman's preliminary calculations showed that the structure shown to him was not consistent with the birefringence of the crystal, and in fact the structure was later modified. This led him on to the idea of using optical and magnetic effects as accessories to X-ray methods for crystal structure determination — concepts which were so elegantly extended and perfected later by his famous student and collaborator K. S. Krishnan.

On his return to India he got his assistants to build an X-ray tube at the Indian Association for the Cultivation of Science. Since the theories of X-ray diffraction by a liquid were quite inadequate at that time, he (with K. R. Ramanathan) developed a satisfactory theory[4] (which A. H. Compton mentions in his book "X-rays and Electrons")[5].

When a pencil of homogenous X-rays passes through a thin layer of liquid and is received on a photographic plate, there appears in addition to the central spot (given by the undeviated beam) diffraction haloes surrounding the centre. The surprise is that the central disk i.e. the first peak of the classical diffraction is absent. It was Raman's interest and insight into the halo phenomenon which provided the explanation. He noted that the X-ray case had to be treated differently from the optical one, since the wavelength of the radiation is of the order of the interatomic distance. The discrete structure of the medium would have to be considered when applying the Einstein-Smoluchowski fluctuations theory. These fluctuations occur over distances varying from that between molecules up to that of the containing vessel. If the density were uniform (i.e. if there are no fluctuations), no scattering would occur, as the interference would be complete. Raman showed that this theory could easily explain why liquids scattered X-rays so little at low angles. At larger angles the variations in the intermolecular distances also have to be computed to explain the spread of the halo. This was done from statistical and thermodynamical considerations again using the Einstein-Smoluchowski ideas as the basis.

THE CLASSICAL DERIVATION OF THE COMPTON EFFECT

In 1924 at the British Association meeting at Toronto, Canada, there was a debate on the recently discovered Compton Effect in which Compton and Raman took part. The discussion is of some interest as it underscores the problems that Physics and Physicists faced in 1924, just before quantum mechanics was formulated. In addition to the X-ray scattering of degraded frequency (the Compton Effect), there is an unmodified secondary radiation. Compton had explained this as due to the whole group of electrons in the atom, scattering conjointly. To this view Raman raised the question:

> "If one electron acting alone can scatter a quantum and also all the Z electrons in the atom acting together, then why do we not observe scattering by two, three or more electrons acting together at a time, with their corresponding fractional Compton shifts in wavelengths? To the alternative explanation of the unmodified scattering given by Professors Compton and Jauncey that it represents the scattering by an electron, which the impinging quantum is unable to detach from the atom, the equally pertinent question may be asked, then why is the intensity of this type of radiation proportional to Z^2 and not Z?."

In 1927 Raman's study of the halo again came to the rescue. Using a simplified atomic model in which electrons are regarded as a gas distributed in a spherical enclosure surrounding the nucleus, he showed that the classical wave principles lead directly to a quantitative theory of the Compton Effect[6].

"The problem is very similar to those which continually arise in such optical problems as the theory of coronae .... The answer to it can be given forth by analogy with the known results in optical cases. The resultant of the Z vibrations can be divided into two parts, *the first part* is entirely determinate, its amplitude being a function of the angle between the primary and secondary rays which is invariable with time. *The second part* is entirely indeterminate so that neither the amplitude nor the phase can be specified at any given time in any given direction and consequently the frequency is also variable. Nevertheless it is possible to specify the statistical expectation of the intensity of this second and highly *fluctuating* type of secondary radiation".

Compton and Allison in their well known book "X-rays in Theory and Experiments"[7] say:

"Raman showed from purely classical considerations that two components must exist ...... "

and go on to give the theory of the scattering of X-rays by an electron cloud*

"the derivation given closely follows Raman[6].

It must be mentioned that Compton too derived the same results independently by a different method two years later[8].

This paper (which was again inspired by his interest in coronae and haloes) is an important one.

He says in it:

"...... our expression represents merely a statistical average of a quantity that *fluctuates with time* ...... the fluctuations with time of the secondary radiation from the atom involve the corresponding fluctuations in the state of the electrical state of the atom ......
To avoid misapprehension it should be made clear that the fluctuations of the atom we are considering are quite different in nature from the fluctuations contemplated in thermodynamics and kinetic theory. We are here concerned with the fluctuations of the atom from its normal condition under the influences of external radiation"[6].

It was this work which convinced him that light radiation can excite molecules and hence there must exist an optical analogue of the Compton Effect — i.e. interaction between light quanta and molecular vibrations.

THE BLUE OF THE SEA

It is an open question as to how much the books one reads in one's youth influences one's activities in later years. In Raman's case, I think there appears to be a connection. I was able to trace two books which were available in Mr R. Chandrasekhara Iyer's (Raman's father's) house which were *certainly* read by Raman when he was between 11 and 13.

"*New Fragments*" by John Tyndall[9] published in 1893 contains an article "about common water" which says

"water of the Lake (of Geneva) is known to be beautifully blue ...... Blue is the natural colour of both water and ice. On the glaciers of Switzerland are found deep shafts and lakes of beautifully blue water. The most striking example of the colour of water is probably that furnished by the Blue Grotto of Capri in the bay of Naples ...... the walls and water of which shimmer forth a magical blue light ...... The bluest of the blue waters are clear and have no detectable suspended impurities."

Popular Lectures on Scientific Subjects by Hermann von Helmholtz[10] contains some magnificent essays — On Goethe's Scientific Re-

* This was probably the first time a detailed method was given for deriving the formula for the atomic scattering factor.

searches. On the Physical Causes of Harmony in Music, On the Relation of Optics to Paintings — essays that must have fascinated young Raman. "On Ice and Glaciers" Helmholtz says

> "In the depths of the crevasses ice is seen of a purity and clearness with which nothing we are acquainted with in the plains can be compared. From its purity it shows a splendid blue colour like that of the sky, only with a greenish hue ...... Their vertical dark blue walls of crystal ice glistening with mositure from the trickling water form one of the most splendid spectacles which Nature can present to us ...... The beautiful blue colour they exhibit is the colour of natural water; liquid water as well as ice is blue, though to an extremely small extent so that the colour is visible in layers from ten to twelve feet thickness".*

Put these along with very different views he read later like the following statement by Lord Rayleigh whose every paper Raman is believed to have read:

> "We must bear in mind that absorption or the proper colour of water cannot manifest itself unless the light traverses a sufficient depth before reaching the eye. In the ocean the depth is of course adequate to develop the colour, but if the water is clear, there is often nothing to send the light back to the observer. Under these circumstances the proper colour cannot be seen. *The much admired dark blue of the deep sea has nothing to do with the colour of water, but simply the blue of the sky seen by reflection*"[11].

Sir Ashutosh Mukherjee (the Vice-Chancellor of Calcutta University) insisted that Raman should definitely go to Oxford for the Universities Congress and this was indeed fortunate. It was on this voyage that he saw the incredible blue of the Mediterranean Sea. He could not believe that the sea could be so blue and so beautiful, nor could he believe that this deep azure was "simply the blue of the sky seen by reflection". Even on boardship he disproved this conjecture of Rayleigh's noting that when the reflection of the sky by the sea is quenched with an analysing nicol prism

> "the colour far from being improverished by suppression of the sky reflection was wonderfully improved ...... it was abundantly clear from the observations that the blue colour of the deep sea is a distinct phenomenon by itself ......"[12]

The rest is history; how, by applying the Einstein-Smoluchowski theory of fluctuations, he established quantitatively that the blue of the sea is due to scattering by molecules — molecular diffraction as he called it; how by laboratory experiments on ice he proved that the blue of the glaciers too arose from molecular scattering. He also showed that molecular scattering was a universal phenomenon in gases, liquids and solids i.e. irrespective of the physical state of the scatterer.

THE SCATTERING OF LIGHT AND THE LIGHT QUANTA

In 1921, even before he voyaged to England, Raman had discussed how molecular movements and molecular vibrations could affect the light scattered by a group of molecules (or a cloud of particles). He had concluded that the movements of the molecules would exhibit themselves as a Doppler shift in the frequency of the incident light, that the shift would be dependent on the angle between the primary and scattered radiation and that it should vanish in the exact forward direction. From his vast experience in acoustical studies, he deduced that if the molecular vibrations are anharmonic, combination tones — sum and difference frequencies — may possibly result, (E. Lommel as early as 1878 considered such a possibility!). However, after his visit to England he seemed to be more concerned with understanding the mechanism of scattering. By

* Raman makes reference to this passage in his paper "Thermal opalescence in crystals and colour of ice in glaciers" *Nature (London)*, 1923, **11**, 13.

imagining molecular scattering to take place in a black body enclosure, Raman convinced himself that Rayleigh Scattering must also take place in a discontinuous manner.

> "We must therefore draw the inference either that the Rayleigh law of scattering is not valid or the molecules do not scatter the radiations incident on them continuously. Since the Rayleigh law of scattering is supported by experiment, at least over a considerable range of wavelengths, it seems more reasonable to accept the latter conclusion, and to infer that the *molecular scattering of light* cannot take place in a continuous manner as contemplated by the classical electrodynamics ...... We are apparently forced to consider the idea that light itself may consist of highly concentrated bundles or quanta of energy travelling through space"[13].

This was strong support of the Einstein idea of the light quantum, a point of view not too popular at that time. As Raman said then

> "though Planck's hypothesis of quantum emission, reinforced as it has been by the success of Bohr's theory of line spectra, has passed into general acceptance, Einstein's idea of light-quanta has apparently been regarded as unnecessarily revolutionary in character"[13].

In 1922, Einstein was awarded the Nobel Prize for his services to theoretical physics and for his work on the photoelectric effect. Pais in Einstein's biography says

> Even when the Einstein photoelectric law was accepted, almost no one but Einstein himself would have anything to do with light quanta!

About two years before his death Nagendra Nath, another of Raman's reputed students told me that in the thirties, [when Raman and Born were great friends(!)] Max Born had told him that he was impressed with Raman's strong advocacy of Einstein's concept of the light quantum in 1921, that he was very pleasantly surprised at Raman's grasping the basic theoretical implications of the Kramers-Heisenberg process, but was truly astounded by Raman's insight in 1921 that Maxwell's field equations would have to be modified to suit the quantum theory!*

In the last chapter of his monograph on "Molecular Diffraction of Light" Raman writes[13]

> "The belief in the validity of Newtonian dynamics as applied to the ultimate particles of matter has, however, received a rude shock from the success of the quantum theory as applied to the theory of specific heats, and there seems no particular reason why we should necessarily cling to Newtonian dynamics, in constructing the mathematical framework of field-equations which form the kernel of Maxwell's theory. Rather, to be consistent, it is necessary that the field-equations should be modified so as to introduce the concept of the quantum of action. In other words, the electrical and magnetic circuits should be conceived not as continously distributed in the field but as discrete units each representing a quantum of action, and possessing an independent existence".

These words were written in 1921. It is interesting that this programme suggested here of quantizing the electromagnetic field was commenced by Dirac in 1928 and by Heisenberg in 1930!

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* Prof. K. R. Ramanathan "whose valuable help in the preparation of the volume and carrying out of the experimental work" has been acknowledged by Raman told me that the manuscript of this monograph *Molecular Diffraction of Light* was submitted to the Press by the last week of December 1921. Raman in his usual manner made innumerable changes in proof. The volume came out by the second week of February 1922. Raman records "his indebtedness to Mr A. C. Ghatak, Superintendent, University Press and his staff "for the quick and efficient manner in which the volume has been printed and got up.

THE DISCOVERY OF THE RAMAN EFFECT

After the publication of this monograph in February 1922, the search started.

> "Bohr's theory has made the idea familiar that the emission or absorption of light from the atom or the expulsion of an electron involves something in the nature of a catastrophic change. If therefore we wish to look for experimental support for Einstein's conception that light itself consists of quantum units, we must consider those optical phenomena in which obviously no such catastrophic change in the atoms or molecules are involved. The molecular diffraction or scattering of light is obviously such a phenomenon"[13].

Even from the beginning, Raman's intuition seems to have told him to look for a change in colour in scattering. He and his collaborators used sunlight and the method of complementary filters to detect this change. Strangely enough, even in the earliest of these experiments he did with K. R. Ramanathan this colour change was noticed but it was attributed to "a weak fluorescence" caused by impurities. At the insistence of Raman, the liquid was purified again and again but the effect persisted.

Said Ramanathan to me later

> "Even in 1923 Prof. Raman refused to believe that this "weak fluorescence" was due to impurities. He said time and again that he felt it was a genuine effect".

The "weak fluorescence" also showed polarization effects but Raman did not, for some strange reason, follow up this important clue as he did later in 1928. In 1924 the "weak fluorescence" was again observed by K. S. Krishnan and in 1925 Raman asked S. Venkateswaran to try to obtain a spectrum of this "weak fluorescence" but no spectrum could be recorded. I have not been able to determine whether this attempt had anything to do with the appearance of the Kramers-Heisenberg paper earlier the same year.

Things came to head in the fall of 1927. Raman, on a holiday in Waltair, worked on and wrote the paper on the classical derivation of the Compton Effect and came back to Calcutta convinced that an "Optical Analogue of the Compton Effect" must exist; and S. Venkateswaran, one of the most diligent of Raman's Collaborators made the remarkable observation that the so-called "fluorescence" in glycerine was strongly polarized. This clearly indicated to Raman that the phenomenon could not be the conventional fluorescence — a point of view he had always taken and for which he was seeking proof.

Venkateswaran was a part-time student who could only work after working hours and on holidays. Raman wanted some one to use the sunlight available all through the day, particularly as he himself had lecturing commitments at the University. And so he pursuaded K. S. Krishnan, the best student he had at that time, to get on to these experiments. Krishnan's diary* says

> "5th February 1928: For the past three or four days I have been doing some experimental work ...... the last experimental work I did was in the summer of. 1926 ......"
> "As Professor says it is not quite healthy for a scientific man to be out of touch with actual experimental facts for any length of time".

Krishnan takes up the problem assigned to him as a dutiful Indian student would, but he is obviously not convinced of the reasons for pursuing these experiments. But within a few days this line of attack led to momentous discoveries".

* Typed extracts from Prof. K. S. Krishnan's diaries were kindly sent to me by Prof. K. R. Ramanathan from which I quote *in extenso*.

"... I took up (at the suggestion of Prof.) the general problem of the 'fluorescence'* of organic vapours, rather than the pressing nature of any specific problem in the subject, awaiting experimental solution which usually draws a man to a new field ...... studied anthracene vapour. It exhibits strong 'fluorescence'; which does not show any polarization ......".

Raman tactfully suggests changing over first to the study of organic liquids, particularly to that of the polarization of the scattered light and verifying his earlier observations.

"Professor has been working with me all the time. Recently Professor has been studying with Mr Venkateswaran the Fluorescence exhibited by many aromatic liquids ...... However in view of the fact that anthracene vapour does not show any polarization, Professor has asked me to verify again his observations on the polarization in some of the liquids ......"

It is remarkable that within two days of Raman's suggestion, Krishnan confirms the observations of Raman and Venkateswaran in many liquids.

"Tuesday, 7th February ...... all pure liquids show fairly intense 'fluorescence' ...... and what is much more interesting all of them are strongly polarized".

Raman verified these observations and wonders why he missed discovering this phenomenon as early as 1923 when Ramanathan had made similar observations.

"He was very much excited and repeated several times that it was an amazing result ...... One after the other, the whole series of liquids were examined and every one of them showed the phenomenon without exception. He wondered how we missed discovering all that five years ago".

Raman then realises that this was the effect he had been looking for, since 1922 (or 1925), a scattering with a modified frequency due to the Kramers-Heisenberg process.

"...... Professor suddenly came to the house (at about 9 p.m.) and called for me. When we went down, we found he was very much excited and had come to tell me that what we had observed this morning must be the Kramers-Heisenberg effect we had been looking for all these days. We therefore agreed to call the effect modified scattering ...... He repeatedly emphasized the exciting nature of the discovery."

Raman then asks Krishnan to go back to the study of vapours to the study of the "modified scattering" in them.

"Thursday, 9th February ...... Tried ether vapour and it was surprising that the modified radiation was conspicuous ...... Professor came from the college at about three ...... and there was enough sunlight to see for himself"

and Raman was in a state of euphoria — a man who had at last come to the end of a trail.

"He ran about the place shouting all the time that it was a first rate discovery, that he was feeling miserable during the lecture because he had to leave the experiment, ... He asked me to call everybody in the place to see the Effect and immediately arranged, in a most dramatic manner, with the mechanics to make arrangements for examining the vapours at high temperatures".

All that remains is to observe it and record it in a spectrograph.

"Tuesday; 28th February. On examining the track with a direct vision spectroscope we found to our great surprise the modified scattering was separated from the

* The quotation marks are mine. The 'fluorescence' referred to (and the 'weak fluorescence' mentioned earlier) is the light seen from the track of the incident beam through a filter complementary to that in the path of the sunlight incident on the liquid or vapour, and is, therefore, of a colour different from that of the Rayleigh Scattered light.

scattering corresponding to the incident light by a dark region ...... This encouraged us to use monochromatic incident light".

I close my article with some remarks the Astrophysicist S. Chandrasekhar of Chicago made recently.

> I have an equally vivid recollection of a day in early March in 1928, when Professor Raman visited our home in Madras on his way to Bangalore where on the 16th of March he was to give the address announcing his discovery of what was soon called the Raman Effect. I remember well his showing slides of the first Raman spectra ever taken and of the state of euphoria he was in. On that occasion some one drew attention to the discovery of the Compton Effect a few years earlier, and Raman responded with 'Ah, but my effect will play a great role for chemistry and molecular structure!' That statement was indeed prophetic. Later during the summer of 1928, I spent two months at the Indian Association for the Cultivation of Science at Raman's laboratory where at that time there were many young men who together with Raman were pursuing the new discovery. Among them were several who were later to become leaders of Indian Science ...... You can imagine what a marvellous experience it must have been for a young man* to have witnessed at such close quarters a group of enthusiastic scientists caught in the wake of a great discovery.

* Chandrasekhar was only 17 then.

4 February 1988

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