

## Preface

# 80<sup>TH</sup> Anniversary of the discovery of the Raman Effect: a celebration

## DISCOVERY

Eighty years ago on 31 March 1928, a letter dated 16 February was published in *Nature* by C. V. Raman (Fig. 1) and K. S. Krishnan with the title 'A New Type of Secondary Radiation'. This letter, which is reproduced in Fig. 2, was the first account in the scientific literature of what soon became known as the Raman Effect. The experiments reported in this letter were based on the use of complementary light filters to detect the presence of modified scattered radiation using focused sunlight as an excitation source and the eye as a detector. Raman and his co-workers soon improved on this rather basic experimental procedure by using a spectroscope to examine the scattered radiation. The first Raman spectrum was reportedly obtained on 28 February 1928. On 29 February the *Associated Press of India* gave the first newspaper report under the heading 'New Theory of Radiation – Prof. Raman's Discovery'; and on 1 March 1928, *The Statesman*, an English language daily of Calcutta, carried an article with the title 'Scattering of Light by Atoms – New Phenomenon – Calcutta Professor's Discovery' which is reproduced in Fig. 3. The paragraph heading 'Two colours from one' is attractive and accurate journalism, but there are several palpable scientific errors in the text. It has to be recognised that Raman's earliest papers were couched in rather general terms and it was only in a letter submitted to *Nature* on 22 March and published on 5 May that a spectrum was presented, although spectra were included in a lecture by Raman in Bangalore on 16 March.

Shortly after the publication of Raman and Krishnan's letter in *Nature*, Landsberg and Mandelstam in Russia reported the observation of light scattering with change of frequency in quartz in a paper in *Die Naturwissenschaften* published on 13 July 1928 and reproduced here in Fig. 4. This paper was submitted on 6 May, one day after the official publication date of Raman and Krishnan's definitive paper. Landsberg and Mandelstam could not have had any information in Moscow about this paper but they did refer to Raman's earlier publications saying that they were unable to decide whether there was any connection with their work because Raman's description was too brief and lacking in detail. The two discoveries must be regarded as quite independent.

Although quantum theory was still in its infancy in the nineteen twenties, the Raman Effect had been predicted by quantum mechanics in two classic papers, one by Smekal<sup>1</sup> published in 1923 and one by Kramers and Heisenberg<sup>2</sup> in 1925.



Figure 1. C. V. Raman.

## DEVELOPMENT

Raman's discovery attracted immediate international interest and was followed up very quickly in many countries. In France Y. Rocard,<sup>3</sup> and separately J. Cabannes<sup>4</sup> published papers in *Comptes Rendus* on the Raman Effect in April 1928. In the United States R. W. Wood<sup>5</sup> quickly followed up Raman's experiments and sent a letter to *Nature* by cable saying that he had verified Raman's work in every particular and describing it as a very beautiful discovery. The wording of Wood's letter is rather awkward because in his cable prepositions and articles were omitted to save money!

In Germany the journal *Die Naturwissenschaften* for 3 August 1928 contained the first review article on the new phenomenon. The author was P. Pringsheim,<sup>6</sup> who proposed that the new effect should be called the Raman Effect.

### A New Type of Secondary Radiation.

If we assume that the X-ray scattering of the 'unmodified' type observed by Prof. Compton corresponds to the normal or average state of the atoms and molecules, while the 'modified' scattering of altered wave-length corresponds to their fluctuations from that state, it would follow that we should expect also in the case of ordinary light two types of scattering, one determined by the normal optical properties of the atoms or molecules, and another representing the effect of their fluctuations from their normal state. It accordingly becomes necessary to test whether this is actually the case. The experiments we have made have confirmed this anticipation, and shown that in every case in which light is scattered by the molecules in dust-free liquids or gases, the diffuse radiation of the ordinary kind, having the same wave-length as the incident beam, is accompanied by a modified scattered radiation of degraded frequency.

The new type of light scattering discovered by us naturally requires very powerful illumination for its observation. In our experiments, a beam of sunlight was converged successively by a telescope objective of 18 cm. aperture and 230 cm. focal length, and by a second lens of 5 cm. focal length. At the focus of the second lens was placed the scattering material, which is either a liquid (carefully purified by repeated distillation *in vacuo*) or its dust-free vapour. To detect the presence of a modified scattered radiation, the method of complementary light-filters was used. A blue-violet filter, when coupled with a yellow-green filter and placed in the incident light, completely extinguished the track of the light through the liquid or vapour. The reappearance of the track when the yellow filter is transferred to a place between it and the observer's eye is proof of the existence of a modified scattered radiation. Spectroscopic confirmation is also available.

Some sixty different common liquids have been examined in this way, and every one of them showed the effect in greater or less degree. That the effect is a true scattering and not a fluorescence is indicated in the first place by its feebleness in comparison with the ordinary scattering, and secondly by its polarisation, which is in many cases quite strong and comparable with the polarisation of the ordinary scattering. The investigation is naturally much more difficult in the case of gases and vapours, owing to the excessive feebleness of the effect. Nevertheless, when the vapour is of sufficient density, for example with ether or amylene, the modified scattering is readily demonstrable.

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

210 Bowbazar Street,  
Calcutta, India,  
Feb. 16.

Figure 2. Facsimile of letter by Raman and Krishnan, published 31 March 1928 in *Nature*, volume 121, page 501.

In the United Kingdom the first Raman publication came from the Clarendon Laboratory, Oxford in the form of a letter to *Nature* from F. A. Lindemann, T. C. Keeley and N. R. Hall.<sup>7</sup> The senior author, F. A. Lindemann, a distinguished physicist, was scientific adviser to Sir Winston Churchill during the 2nd World War and later served in his post-war Cabinet. He was created Lord Cherwell (later Viscount Cherwell) for his services.

In Austria K. W. F. Kohlrausch began a long series of experimental studies of the Raman Effect at the University of Graz, as a result of reading R. W. Wood's paper. A first experiment using an old mercury lamp as a source, freshly distilled carbon tetrachloride in a round-bottomed flask as a sample, a Zeiss spectrograph and a photographic plate as detector gave a satisfactory Raman spectrum of carbon tetrachloride. Encouraged by this result Kohlrausch and his co-workers set about recording Raman spectra with great enthusiasm. A. Dadiou, one of the co-workers at Graz, has described the atmosphere in the laboratory:

*From then on, the Raman lamp burned day and night. We slept in turns in the Institute, I myself distilled and cleaned until midnight the liquids we meant to analyse, made more exposures and measured the spectra. Kohlrausch saw to the marking of the lines, worked out the resulting Raman spectra and tried to interpret their structure. After two months we sent our first paper<sup>8</sup> to the Austrian Academy of Science. Another five papers right across organic chemistry followed up to the end of 1929.*

For some 15 years or so Kohlrausch's group at Graz made extensive studies of the Raman spectra of an extraordinarily wide range of organic compounds. The results of these painstaking and valuable studies were published in three classic volumes on 'Der Smekal Raman-effekt' the first of which appeared in 1931.<sup>9</sup>

There were also a number of notable contributions from Italy in this period. F. Rasetti published a series of classical papers dealing with the pure rotational Raman spectra of gases and the associated selection rules. In an important paper published in 1930 in *Zeitschrift für Physik* Rasetti<sup>10</sup> reported the pure rotational Raman spectra of gaseous nitrogen and oxygen. He showed how a nuclear spin of 1 for <sup>14</sup>N produces an intensity alternation with the rotational lines of even and odd J values having intensities in the ratio 2:1. Similarly, a nuclear spin of zero for <sup>16</sup>O leads to the absence of rotational lines with even J values. He also reported in *Nature*<sup>11</sup> the first observation of an electronic transition in a Raman spectrum. Doublets in the Raman spectrum of NO with a separation of 124 cm<sup>-1</sup> were assigned to transitions between the two electronic states <sup>2</sup>Π<sub>1/2</sub> and <sup>2</sup>Π<sub>3/2</sub>. The Italian scientists E. Amaldi, A. Carelli, E. Fermi and E. Segré made important contributions to the theory of the Raman Effect. In Bologna, G. B. Bonino appreciated that the Raman Effect was a valuable tool for research relating to organic molecules. He has recalled the excitement he felt when one day in 1928 he found Raman and Krishnan's paper in his university library. He quickly realised that a Raman spectrum would be easier to obtain than an infra-red spectrum using the techniques then available.

Laboratories in many other European countries began to apply Raman spectroscopy to a variety of problems but, because of limitation of space, only brief citations can

# SCATTERING LIGHT BY ATOMS

## NEW PHENOMENON

### CALCUTTA PROFESSOR'S DISCOVERY

PROF. C. V. Raman, F.R.S., of the Calcutta University, has made a discovery which promises to be of fundamental significance to physics.

Prof. A. H. Compton, of Chicago University, was recently awarded the Nobel Prize for his discovery of the remarkable transformation which X-rays undergo when they are scattered by

atoms. Shortly after the publication of Prof. Compton's discovery, other experiments sought to find whether similar transformation occurs also when ordinary light is scattered by matter and they reported definitely negative results.

Prof. Raman, with his research associates, took up this question afresh, and his experiments have disclosed a new kind of radiation from atoms excited by light. The new phenomenon exhibits features even more startling than those discovered by Prof. Compton with X-rays.

#### TWO COLOURS FROM ONE

The principal feature observed is that when matter is excited by light of one colour, the atoms contained in

it emit light of two colours, one of which is different from the exciting colour and is lower down the spectrum.

The astonishing thing is that the altered colour is quite independent of the nature of the substance used. It changes, however, with the colour of the exciting radiation, and if the latter gives a sharp line in the spectrum, the second colour also appears as a second sharp-line. There is in addition a diffuse radiation spread over a considerable range of the spectrum.

Prof. Raman's discovery is almost incredible as it is not indicated by existing theories of radiation. He will deliver a lecture demonstrating these phenomena at Bangalore on March 16.

Figure 3. Facsimile of announcement in the Calcutta newspaper *The Statesman*, 1 March 1928.

be given. In Switzerland R. Bar<sup>12</sup> studied Raman spectra obtained from powdered crystals. In Denmark Langseth<sup>13</sup> studied the relation between Raman spectra and ultraviolet absorption. In Poland W. Czapska<sup>14</sup> reported the Raman spectra of *para*, *ortho* and *meta* xylenes. In Belgium Manneback<sup>15</sup> discussed the intensities of Raman lines and in Norway Bjorn Trumphy<sup>16</sup> was active in the determination of the degree of polarization of Raman scattering and deducing molecular symmetry therefrom. Harm Gerding<sup>17</sup> in The Netherlands used Raman spectroscopy to study sulphur trioxide in the liquid state, in solution in sulphur dioxide and in the gaseous state. The first Japanese paper on Raman spectroscopy came from M. Kimura and Y. Uchida<sup>18</sup> with the enigmatic title 'The Raman Spectra of Certain Substances.'

On 20 December 1928 J. C. McLennan and J. H. McLeod<sup>19</sup> of the Physics Department, University of Toronto, Canada sent a letter to *Nature* entitled 'the Raman effect with liquid Oxygen, Nitrogen and Hydrogen'. This was published on 2 February 1929.

The results of McLennan and McLeod were of very great significance. They gave direct experimental evidence that: (i) the vibrations of homopolar molecules which are infrared inactive can be observed in the Raman Effect; (ii) two quantum rotational transitions occur in Raman scattering; (iii) hydrogen at low temperatures must be regarded as a mixture of two effectively distinct sets of molecules, symmetrical and antisymmetrical. This work formed one part of the Ph.D thesis of J. H. McLeod submitted to the University of Toronto in November 1929. The other part of the thesis was concerned with the nature of the auroral green spectral line, to which McLeod also made a significant contribution. His outstanding Ph.D thesis is further remarkable for its brevity, consisting of just 21 pages! Pages 13–21 deal with the Raman work including detailed suggestions for further work. A page from this remarkable thesis is reproduced in Fig. 5.

A General Discussion of the Faraday Society<sup>20</sup> was held on 25 September 1929 in the Physics Department of the University of Bristol. The cost, including full board, was £1.25 for members and £1.50 for non-members! It was attended by many eminent spectroscopists. Raman presented a paper entitled 'Investigation of molecular structure by light scattering' to open the section devoted to the Raman Effect; other contributors to this section included J. Cabannes, R. W. Wood, P. Daure, A. C. Menzies and J. C. McLennan.

Dr. G. Herzberg provided the author with the following reminiscences of this meeting;

*'I met Raman for the first time in 1929 at the General Discussion of the Faraday Society on Molecular Spectra and Molecular Structure. It was a very memorable meeting largely because of the presence of Raman. For me it was the first time that I attended a meeting held in the English language. The first speaker was O. W. Richardson, who unfortunately mumbled and made it quite impossible for me to understand him. The second speaker was Raman. He spoke clearly and lucidly in beautiful English that even I could understand and his enthusiasm was certainly infectious. Just shortly preceding this meeting Heitler and I had discussed the importance of Rasetti's result about the intensity alternation in the Raman spectrum of nitrogen for nuclear theory and Raman showed considerable interest in and understanding of our result.'*

Dr. A. C. Menzies recalled that Raman, during his stay at Bristol, regularly enquired of the hotel porter if there was a cable for him from Stockholm! In fact Raman had to wait until 1930 for his Nobel Prize.

The career of Raman is particularly interesting and helps to set in perspective his discovery of the Raman Effect. Chandrasekhara Venkata Raman was born at Trichinopoly in South India on 7 November 1888. His family moved to

**Eine neue Erscheinung bei der Lichtzerstreuung in Krystallen.**

Bei dem Studium der molekularen Lichtzerstreuung in festen Körpern, welches zur Klärung der Frage vorgenommen wurde, ob dabei eine Wellenlängenänderung stattfindet, was man nach der DEBYESchen Theorie der spezifischen Wärme vermuten kann, haben wir eine neue Erscheinung gefunden, die, wie es uns scheint, ein bedeutendes Interesse beansprucht.

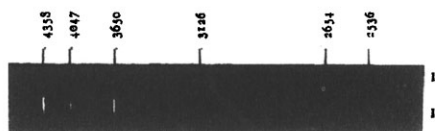
Diese Erscheinung besteht in der Wellenlängenänderung, welche aber von anderer Größenordnung ist, als die von uns erwartete und welche einen ganz anderen Ursprung hat.

Ein intensives Lichtbündel von einer Quecksilberquarzlampe wurde durch ein Quarzkrystall gesandt, und das senkrecht zu dem primären Bündel zerstreute Licht wurde mittels eines Quarzspektrographen aufgenommen. Die gewöhnlichen Maßregeln gegen fremdes Licht wurden getroffen<sup>1</sup>. Als Vergleichsspektrum diente eine Aufnahme des von schwarzem Samtreflektierten Lichtes. Expositionszeit von 2–14 Stunden. Die Versuche wurden mit zwei verschiedenen Quarzstücken ausgeführt. Es erwies sich, daß auf allen Spektrogrammen alle Quecksilberlinien von je einem deutlich ausgeprägten Trabanten von etwas größerer Wellenlänge begleitet waren, und außerdem noch bei jeder Linie zwei oder drei weniger ausgeprägte andeutungsweise zum Vorschein kamen. In dem Vergleichsspektrum war keine Spur dieser Trabanten zu sehen. Fig. 1 gibt eines der Spektrogramme wieder. Die angenäherten Ausmessungen der Spektrogramme zeigen für den stärkeren Trabanten folgende Wellenlängenänderung:

Tabella 1.

$\lambda$ in Å	$\Delta\lambda$	
	beobachtet	berechnet
2536	ca. 30	30,8
3126	.. 47	47,0
3650	.. 63	64,0

Wir haben verschiedene Kontrollversuche angestellt, um festzustellen, daß die beobachteten Linien nicht von einem zufälligen falschen Licht herrühren. Der folgende Versuch scheint uns entscheidend zu sein. Zwischen dem zerstreuenden Quarzkrystall und dem Spektrographspalt wurde ein Quarzgefäß mit Quecksilberdampf eingeschaltet, welcher die Linie 2536 Ång-



I. Das Spektrum des zerstreuten Lichtes.  
II. Das Vergleichsspektrum.

ström vollständig absorbiert. Auf dem Spektrogramm haben wir diese Linie nicht mehr erhalten, wohl aber ihre Trabanten. Das beweist sicher, daß diese Trabanten wirklich eine andere Wellenlänge haben als die Grundlinie.

Wir halten es für verfrüht, schon jetzt eine definitive Deutung der geschilderten Erscheinung zu geben. Eine der möglichen theoretischen Deutungen besteht vielleicht in folgendem: Bei der Zerstreuung des Lichtes können einige eigene ultrarote Frequenzen des Quarzes auf Kosten der Energie des zerstreuten Lichtes angeregt werden und dadurch würde die Energie der zerstreuten Quanten und folglich ihre Frequenz um die Größe der entsprechenden infraroten Quanten abnehmen. Geht man dabei von der Frequenz, welche der Wellenlänge  $\lambda = 20,7 \mu^1$  entspricht, aus, so erhält man eine gute Übereinstimmung zwischen den berechneten und tatsächlich beobachteten Werten (s. Tab. 1).

Ob und wie weit die von uns beobachtete Erscheinung mit der von RAMAN<sup>2</sup> erst kürzlich beschriebenen im Zusammenhang steht, können wir zur Zeit noch nicht beurteilen, weil seine Schilderung zu summarisch ist.

Moskau, Institut für theoretische Physik der I. Universität, den 6. Mai 1928.

G. LANDSBERG. L. MANDELSTAM.

Anmerkung bei der Korrektur. Wir haben inzwischen das im Kalkspat zerstreute Licht untersucht und dieselbe Erscheinung beobachtet. Die Wellenlängenänderung ist hier größer als im Quarz. Sie würde einer Infrarotenfrequenz von  $\lambda = 9,1 \mu$  entsprechen.

<sup>1</sup> G. LANDSBERG, Zeitschr. f. Physik 43, 773. 1927; 45, 442. 1927.

Figure 4. Facsimile of paper by Landsberg and Mandelstam published 13 July 1928 in *Die Naturwissenschaften* volume 16, page 557.

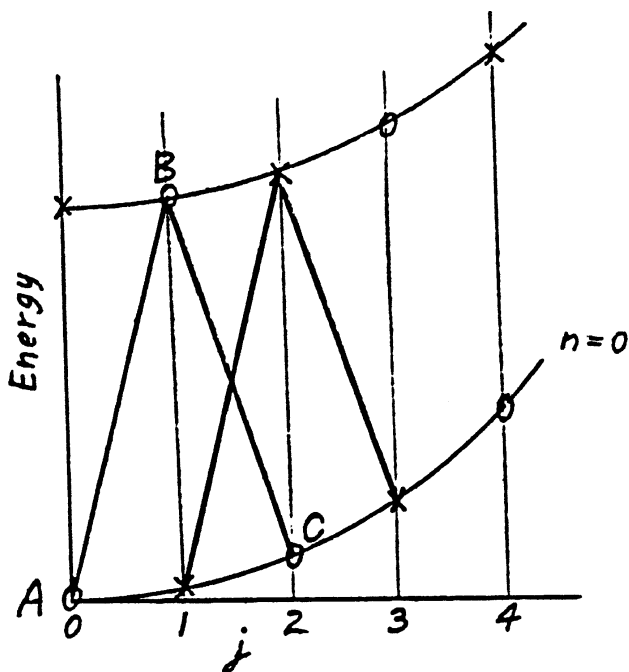


Figure 5. Facsimile of page 15 of the Ph.D. thesis of J. H. McLeod submitted to the University of Toronto, Canada in November 1929.

Visagapatam on the East Coast of India where he went to the local High School. In 1903 he entered the Presidency College of Madras.

Even as a student at the Presidency College, Raman undertook original investigations in acoustics and optics. His first paper<sup>21</sup> entitled 'Unsymmetrical diffraction bands due to a rectangular aperture' was published in the *Philosophical Magazine* (London) in 1906. This was followed early in 1907 by a note<sup>22</sup> in *Nature* on 'Newton's rings in polarized light'. Both these papers were published in Raman's eighteenth year.

After graduating he spent ten years in the Indian Finance Department, Calcutta, as Assistant Accountant General, but maintained an active interest in science, carrying out research in his spare time at the Indian Association for the Cultivation of Science, a privately endowed institution in Calcutta, founded in 1876. During this period he contributed some 30 original papers in physics to *Nature*, the *Philosophical Magazine* and *Physical Review*.

In 1917 he joined Calcutta University as the first Palit Professor of Physics but continued his connection with the Indian Association for the Cultivation of Science of which he became the Honorary Secretary in 1919.

At Calcutta he was active in research in many areas of physics including the theory of musical instruments, diffraction, colours, interference, colloids, light scattering, X-rays, magnetism and magneto-optics. He rapidly established himself as a physicist of international renown and was elected Fellow of the Royal Society of London in 1924.

Raman travelled widely in the 1920s attending international congresses and lecturing at universities in many countries. His zest for travel in general and Mediterranean voyages in particular led to his becoming interested in the phenomenon of light scattering and so paved the way for the discovery of the effect which bears his name.

Raman received many honours, including a knighthood in 1929 and the Nobel Prize for Physics in 1930. He left Calcutta in 1933 to become Director of the Indian Institute of Science at Bangalore. Later he was able to build an Institute of his own in Bangalore, now known as the Raman Research Institute. Here he lived and worked actively until his death on 21 November 1970. His mortal remains were cremated in the beautiful gardens of his Institute.

There are many comments on record about Raman which illustrate his great talents, his human qualities and his complex personality:

*'The Raman effect must rank among the best three or four discoveries in experimental physics in the last decade.'*  
(Lord Rutherford, 1930)

*'C. V. Raman was the first to recognize and demonstrate that the energy of a photon can undergo partial transformation within matter. I still recall vividly the deep impression that this discovery made on all of us. . . .'* (A. Einstein)

*'He was richly endowed with a child-like sense of wonder at the unknown and not understood facets of nature. . . .'*  
(S. Bhagavantam)

*'His exuberance was infectious. . . .'* (S. Ramaseshan)

*'I must frankly say I like him (Raman) very much, in spite of his all too human drawbacks, his conceit and his naiveté and therefore his disarming way of bringing himself into the light. I do not take these too seriously as I am finding (on the other hand) a genuine devotion to his work and to the Institute. He is an excellent physicist, full of optimism and activity and besides being interested in everything in the world.'* (Max Born in a letter to Lord Rutherford, 1936)

But perhaps Raman himself should have the last word: *'The essence of science is independent thinking, hard work and not equipment. When I got my Nobel Prize I had spent hardly 200 rupees on my equipment.'*

## FURTHER READING

More detailed documentation will be found in my review article<sup>23</sup> entitled 'Early history of the Raman effect.' It is

hoped that a further review article covering the subsequent history of Raman spectroscopy up to ca. 1980 will appear in this journal towards the end of 2008.

## PERSONAL NOTE

I recorded photographically my first Raman spectrum (of CCl<sub>4</sub>) in October 1946 in the Sir Leoline Jenkins Laboratories of Jesus College, Oxford. The apparatus was very similar to that used by Raman to obtain his first Raman spectra. It consisted of a small Hilger constant deviation spectrograph of classical mahogany and brass construction. The irradiating light source consisted of a commercial Hanovia quartz mercury sun lamp mounted at one focus of an elliptical reflector with the sample cell mounted at the other focus. The elliptical reflector was made by the college carpenter and chromium plated at the Radiator Works of the Morris Oxford Car Plant.

My research supervisor was Dr. L. A. Woodward, Chemistry Fellow of Jesus College. After graduating in Chemistry in Oxford he went on a Government Scholarship to Leipzig to work for a Ph.D. under the fertile direction of Professor P. Debye. He chose to work on the newly discovered Raman Effect. His first published paper<sup>24</sup> was on the Raman Effect of nitric acid in solution and appeared in *Nature* in 1930. In 1931 he was awarded his doctorate, *summa cum laude*. The title of his thesis was 'Raman spectra of electrolyte solutions'.

## Acknowledgements

I acknowledge with gratitude the kindness of the many friends and colleagues who have contributed to my archives over the years. I thank Emeritus Professor Michel Gilbert of Liège University, Belgium, who provided the splendid photograph of C. V. Raman taken when he visited Liège. I am also much indebted to my wife, Moira, whose scholarly help and advice have been of great assistance and encouragement in the preparation of this article.

**D. A. Long**  
University of Bradford, UK

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